

FORM PTO-1390 (REV 11-96)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER	
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				OGOH:079	
INTERNATIONAL APPLICATION NO. PCT/JP00/06971		INTERNATIONAL FILING DATE 5 October 2000		U.S. APPLICATION NUMBER (see 37 CFR 1.5) <b>09/857424</b>	
TITLE OF INVENTION LIGHT EMITTING ELEMENT AND PRODUCING METHOD THEREOF, AND DISPLAY DEVICE AND LIGHTING DEVICE USING THE SAME				PRIORITY DATE CLAIMED 5 October 1999 21 October 1999	
APPLICANT(S) FOR DO/EO/US Mikiko MATSUO; Tetsuya SATO; Hisanori SUGIURA; Hitoshi HISADA					
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:					
<ol style="list-style-type: none"> <li><input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li><input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li><input checked="" type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f) at any time <del>rather than delay</del> examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).</li> <li><input type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</li> <li><input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))           <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</li> </ol> </li> <li><input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)).</li> <li><input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))           <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input type="checkbox"/> have been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</li> <li>d. <input type="checkbox"/> have not been made and will not be made.</li> </ol> </li> <li><input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</li> <li><input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</li> <li><input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</li> </ol>					
Items 11. to 16. below concern document(s) or information included:					
<ol style="list-style-type: none"> <li><input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.</li> <li><input checked="" type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</li> <li><input type="checkbox"/> A <b>FIRST</b> preliminary amendment.</li> <li><input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.</li> <li><input type="checkbox"/> A substitute specification.</li> <li><input type="checkbox"/> A change of power of attorney and/or address letter.</li> <li><input checked="" type="checkbox"/> Other items or information: International Application Cover Page; International Search Report; PCT Request; Forms PCT/IB/301; 304 and 308.</li> </ol>					

U.S. APPLICATION NO. (if known) <b>09/857424</b>	INTERNATIONAL APPLICATION NO PCT/JP00/06971	ATTORNEYS DOCKET NUMBER OGOH-079	
17. <input checked="" type="checkbox"/> The following fees are submitted:		<b>CALCULATIONS PTO USE ONLY</b>	
<b>BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5) ) :</b>			
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO ..... \$1,000.00			
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... \$860.00			
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... \$760.00			
International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... \$690.00			
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) ..... \$100.00			
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>		\$ 860.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).		\$	
<b>CLAIMS</b>	<b>NUMBER FILED</b>	<b>NUMBER EXTRA</b>	<b>RATE</b>
Total claims	62 - 20 =	42	X \$18.00 \$ 756.00
Independent claims	21 - 3 =	18	X \$80.00 \$ 1,440.00
<b>MULTIPLE DEPENDENT CLAIM(S) (if applicable)</b>		+ \$270.00 \$	
<b>TOTAL OF ABOVE CALCULATIONS =</b>		\$ 3,056.00	
Reduction of 1/2 for filing by small entity, if applicable. A Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28).		\$	
<b>SUBTOTAL =</b>		\$ 3,056.00	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).		\$	
<b>TOTAL NATIONAL FEE =</b>		\$ 3,056.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property		+ \$ 40.00	
<b>TOTAL FEES ENCLOSED =</b>		\$ 3,096.00	
		<b>Amount to be:</b>	\$
		<b>refunded</b>	\$
		<b>charged</b>	\$
a. <input checked="" type="checkbox"/> A check in the amount of \$ 3,096.00 to cover the above fees is enclosed. CK# <u>14064</u>			
b. <input type="checkbox"/> Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.			
c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>16-0331</u> . A duplicate copy of this sheet is enclosed.			
<b>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.</b>			
SEND ALL CORRESPONDENCE TO: Roger W. Parkhurst PARKHURST & WENDEL, L.L.P. 1421 Prince St., Ste. 210 Alexandria, VA 22314-2805 Tel: (703) 739-0220		 SIGNATURE <u>Roger W. Parkhurst</u> NAME 25,177 REGISTRATION NUMBER	

LIGHT EMITTING ELEMENT AND PRODUCING METHOD THEREOF,  
AND DISPLAY DEVICE AND LIGHTING DEVICE USING THE SAME

Technical Field

5 The present invention relates to a light-emitting device used as a flat light source and a flat display.

Background Art

Electric field light-emitting devices, which have self-luminous property and thus high visibility and excellent display capability and enable high 10 speed response and low-profile, are now attracting the attention as displays for flat displays and the like.

Among others, an organic EL device, using organic compound as phosphor, has characteristic features, as compared with inorganic EL device, that it can be driven at a low voltage, that it can easily produce an enlarged 15 area, and that it can easily produce a desired luminous color by an appropriate choice of coloring matter and, accordingly, the organic EL device is now vigorously developing as a next-generation display.

The EL device using the organic phosphor generates blue luminescence for example, through the application of a voltage of 30 volts to an 20 anthracene evaporated film having thickness of 1 m or less (Thin Solid Films, 94(1982) 171). However, this device fails to produce sufficient luminance even when a high voltage is applied thereto, so that it is required to be further improved to have higher luminous efficiency.

In regard to this, Tang et al. teaches that transparent electrodes 25 (anode), a hole transport layer, an emission region having electron transport

capabilities, cathode using metal of low work function are laminated for reduction in voltage and improvement in luminous efficiency, to thereby produce luminance of 1,000 cd/m<sup>2</sup> through the application of a voltage of 10V or less (Appl. Phys. Lett. 51 (1987) 913). Used as the phosphor is tris 5 (8-quinolinolato) aluminum complex (hereinafter it is referred to as "Alq"). The Alq is an excellent luminophor having both high luminous efficiency and high electron transport capability.

Further, a device having a three-ply structure wherein an emission region is sandwiched between a hole transport layer and an electron 10 transport layer and a device that obtains luminescence from coloring matter (coumarin derivative or fluorescent dye, such as DCM1, used to the Alq) doped in the emission region is reported (Jpn. J. Appl. Phys., 27 (1988) L269 and J. Appl. Phys., 65(1989) 3610). The report says that it is discovered that an adequate choice of the coloring matter can allow luminous color to 15 change and can produce improved luminous efficiency, as compared with the non-doped one.

In the devices constructed above, all layers are formed by dry process such as a vacuum evaporation method. On the other hand, the method has been proposed of producing the device in the so-called wet deposition process 20 such as a spin coat method and a cast method (Japanese Laid-open (unexamined) Patent Publications No. Hei 3-790 and No. Hei 3-171590).

In this method, at least one material for forming the hole transport layer, the electron transport layer and the emission region is dissolved in appropriate solvent, together with polymer binder. Then, that solution is 25 applied onto the electrode to form the emission region and, thereafter, a

further electrode is formed on the emission region in the evaporation method or a like method. Hereinafter, the organic light-emitting device thus produced is called "polymer dispersed light-emitting device", in contrast to the conventional laminated light-emitting device.

5        When comparing with the organic light-emitting device produced by the dry process, the polymer dispersed light-emitting device has the following advantages.

(1) Even the material for which it is hard to be deposited in the dry process such as evaporation can be used;

10      (2) Even the doping of minute amounts for which it is hard to be controlled in the dry process can be performed with ease;

(3) An enlarged area can be produced with ease;

(4) The device can be produced at low costs;

(5) Simultaneous luminescence from luminous molecules is enabled  
15      with ease through the introduction of a number of luminous molecules  
(white luminescence is enabled); and

(6) In the conventional laminated light-emitting device, the layers are each in the amorphous state. In contrast to this, in the polymer dispersed light-emitting device, the materials are dispersed in the polymer binder.

20      Accordingly, the latter is thermally stable as compared with the former.

The conventional polymer dispersed light-emitting devices comprise the emission region in which perinone derivative or tris (8-quinolinolato) aluminum used as the luminous molecules is dispersed in polyvinyl carbazol or the emission region in which tris (8-quinolinolato) aluminum and  
25      tetraphenyl benzidine used as the luminous molecules are dispersed in

polycarbonate (Japanese Laid-open (unexamined) Patent Publications No. Hei 3-790, No. Hei 3-171590).

(First problem)

The polymer dispersed light-emitting device has the advantages 5 mentioned above, while on the other hand, it has the disadvantage that it is low in luminous efficiency, as compared with the conventional laminated light-emitting device.

Specifically, in the laminated light-emitting device, holes are injected from the anode to the hole transport layer, and electrons are injected from 10 the cathode to the emission region of the electron transport capability or the electron transport layer. When these holes and electrons are recombined in the emission region, excitons are formed, and when the excitons are caused to transition to the ground state, the emission takes place. It is to be noted that the electron transport and the hole transport are functionally separated 15 from each other, so that the recombination of the electron and hole takes place only in the vicinity of an interface between the adjoining layers. This can produce the efficient production of the excitons and thus improved luminous efficiency.

Further, through an appropriate choice of the material of the layers 20 adjoining the respective electrodes to minimize injection barriers between the anode and the cathode, the injection of the hole and the electron can be facilitated, and as such can allow the drive at a low voltage.

In contrast to this, the polymer dispersed light-emitting device mainly comprises a monolayer, so that the recombination of the hole and electron 25 and the production of the excitons do not take place locally, differently from

the laminated light-emitting device mentioned above. In addition, the barriers for the hole and the electron to be injected from the electrodes are high. These facts make it difficult to improve luminous efficiency.

Thus, the emission site is dispersed over the whole emission region, so  
5 that it is hard to balance the injection of the hole and electron and the transport of the same. As a result of this, the probability of the recombination reduces, such that sufficient luminous efficiency is not produced. This indicates that collection of the recombination region of the hole and electron into a specific region leads up to improvement in luminous  
10 efficiency.

Though the method of laminating the functionally separated layers is of effective, as mentioned above, in a polymeric system formed by coating, solvent contained in polymer solution of a second layer to be laminated over a first deposited layer must be chosen to prevent the first deposited layer  
15 from being dissolved in that solvent.

In addition, as the laminated layers increase in number, the need to choose the solvent and the need to choose the material soluble in that selected solvent arise increasingly. This arises the problem that the range of choice for the material is further narrowed, thus hindering the effectively  
20 improved efficiency.

(Second problem)

Further, the polymer dispersed light-emitting device involves the problem that when a color panel is produced, it is difficult to do the patterning (distribution of application of color). When the color panel is  
25 produced in the dry process such as the vacuum evaporation method,

elements of color can be formed in desired positions by an evaporation mask being set on a substrate. In contrast to this, in the wet deposition such as the spin coat method or the cast method, since the emission region is formed over the whole area of the substrate, the above-mentioned patterning cannot

5 be performed.

In regard to this, the patterning using an ink-jet method has been proposed (e.g. Japanese Laid-open (unexamined) Patent Publication No. Hei 10-12377). This proposes that material of the emission region including polymer or the precursor of polymer is discharged from nozzles by the

10 ink-jet method to form a desired pattern.

However, the patterning using the conventional ink-jet method involves the problem that polymer solution or equivalent to be coated is high in viscosity, so that the nozzles of the ink head are easily plugged up and, accordingly, it is difficult to form a micropattern.

15 To solve the problems mentioned above, the present invention has been made. The present invention provides an organic light-emitting device capable of producing highly improved luminous efficiency and at the same time facilitating the patterning even in the polymer dispersed organic light-emitting device and the producing method thereof.

20

### Disclosure of the Invention

In the light of the current situation, a group of inventions have been made. The object of the invention is to provide a light-emitting device having high luminous efficiency.

25 Although the group of inventions is based on the same or similar

conception, since they are realized by way of different embodiments, the inventive groups are divided into the first inventive group and the second inventive group on the basis of their respective relevance in the specification.

In the following, the contents will be explained in order for each division

5 (each inventive group).

(First inventive group)

After having devoted themselves to the studies in order to attain the object, the inventors of this application have found that in producing a polymer dispersed light-emitting device, a polymer layer is first formed and  
10 then luminous molecules, or luminous molecules and charge transport are penetrated in the polymer layer thus formed, thereby producing a highly improved luminous efficiency and at the same time facilitating the patterning.

Specifically, a first aspect of the invention is directed to a  
15 light-emitting device having an emission region between an anode and a cathode, wherein the emission region comprises material contributable to emission and a medium for containing the material, and wherein the material contributable to the emission has a substantially successive distribution of concentration from the anode side of the emission region  
20 toward the cathode side thereof.

A second aspect of the invention is directed to the light-emitting device as set forth in the first aspect of the invention, wherein the material contributable to the emission exhibits the distribution of concentration according to which any one of the anode side of the emission region and the  
25 cathode side thereof is higher in concentration than the other, and the

concentration reduces from the one side toward the other side successively.

With this constitution, in a region where the material contributable to the emission in the emission region is high in concentration, the hole injected from the anode into the emission region and the electron injected from the cathode into the emission region are recombined. Thus, the recombination region of the hole and the electron is collected, so that the recombination efficiency of the electron and the hole is increased and thus the luminous efficiency is improved.

The terminology "material contributable to emission" is intended to include, for example, material that form organic binder, charge transport material and dimmer, excimer, or exciplex to obtain emission therefrom, in addition to the luminous molecules that emit light by the injection of electric charge as shown in Embodiments as will be mentioned later. It is noted here that the terminology of the dimmer means material combined with the organic binder or the charge transport material in the ground state and the terminology of the excimer and the exciplex means material caused to react with the organic binder or the charge transport material in the excitation state caused by the injection of the charge.

A third aspect of the invention is directed to the light-emitting device as set forth in the first aspect of the invention, wherein the emission region further comprises charge transport material.

The charge transport capabilities in the emission region are further improved and thus the recombination efficiency of the electron and the hole is improved.

A fourth aspect of the invention is directed to the light-emitting device

as set forth in the third aspect of the invention, wherein the charge transport material has a substantially successive distribution of concentration from the anode side of the emission region toward the cathode side thereof.

5       With this constitution, a region large in hole transport capability and a region large in electron transport capability are formed in the emission region, as viewed in a direction extending from the anode side to the cathode side (layer thickness direction), so that the recombination efficiency of the electron and the hole is improved.

10      A fifth aspect of the invention is directed to a light-emitting device having a charge transport region between an anode and a cathode, wherein the charge transport region comprises charge transport material and a medium for containing the charge transport material, and wherein the charge transport material has a substantially successive distribution of 15 concentration from the cathode side of the charge transport region toward the anode side thereof.

With this constitution, a region large in hole transport capability and a region large in electron transport capability are formed in the emission region, so that the recombination efficiency of the electron and the hole is 20 improved. In the constitution above, the charge transport region has luminescent properties and thus doubles as the emission region.

A sixth aspect of the invention is directed to the light-emitting device as set forth in the first aspect of the invention, wherein the emission region includes a region where the material contributable to the emission is not 25 present.

With this constitution, a region different in carrier transport capability is formed in the emission region, so that the recombination efficiency of the electron and the hole is further improved and thus the luminance efficiency is improved.

5 A seventh aspect of the invention is directed to the light-emitting device as set forth in the first aspect of the invention, wherein a part of the emission region that exhibits the maximum concentration of the material contributable to the emission is away from the anode and the cathode.

The reason that the part of the emission region that exhibits the  
10 maximum concentration of the material contributable to the emission is away from the both electrodes is that when the material contributable to the emission is close to the anode or the cathode, there is the possibility that the material contributable to the emission may be extinguished without emitting. The part of the emission region that exhibits the maximum  
15 concentration of the material contributable to the emission is preferable at a substantially immediate position between the anode and the cathode.

An eighth aspect of the invention is directed to the light-emitting device as set forth in the fifth aspect of the invention, wherein the charge transport region includes a region where the charge transport material is  
20 not present.

With this constitution, in the boundary between the region where the charge transport material is present and the region where the charge transport material is not present a region, the recombination efficiency of the electron injected from the cathode and the hole injected from the anode  
25 is further improved and thus the luminance efficiency is improved.

A ninth aspect of the invention is directed to the light-emitting device as set forth in the fifth aspect of the invention, wherein a part of the charge transport region that exhibits the maximum concentration of the charge transport material is away from the anode and the cathode.

5 A tenth aspect of the invention is directed to the lighting device using the light-emitting device as set forth in the first aspect of the invention.

With this constitution, the lighting device having improved luminance efficiency can be provided.

An 11th aspect of the invention is directed to a light-emitting device  
10 having an emission region between an anode and a cathode, wherein the emission region comprises material contributable to emission and a medium for containing the material, and wherein the material contributable to the emission has a distribution of concentration that reduces substantially successively in a direction parallel to a surface of the cathode and a surface  
15 of the anode from a substantially center of the emission region toward a periphery thereof.

A 12th aspect of the invention is directed to the light-emitting device as set forth in the 11th aspect of the invention, wherein there are provided a number of materials contributable to the emissions and are adjacently  
20 arranged in a direction parallel to a surface of the cathode and a surface of the anode, and wherein the materials contributable to the emissions are different from each other in luminous color.

With this constitution, since the concentration is low in regions in the vicinity of the boundaries between the materials contributable to the  
25 emission, the materials contributable to the emission are prevented from

being mixed with each other. Thus, the light-emitting device that provides excellent full color display capability with little color mixture is produced.

A 13th aspect of the invention is directed to the light-emitting device as set forth in the 11th aspect of the invention, wherein the emission region 5 further comprises charge transport material.

A 14th aspect of the invention is directed to the light-emitting device as set forth in the 13th aspect of the invention, wherein the charge transport material has a distribution of concentration that reduces in the direction parallel to the surface of the cathode and the surface of the anode 10 from the substantially center of the emission region toward the periphery thereof.

A 15th aspect of the invention is directed to the light-emitting device as set forth in the 11th aspect of the invention, wherein the material contributable to the emission has a substantially successive distribution of 15 concentration from the cathode side of the emission region toward the anode side thereof.

With this constitution, since the material contributable to the emission is made to have the distribution of concentration so that the recombination region of the hole and the electron is collected, the recombination efficiency 20 of the electron and the hole is improved and thus the luminance efficiency is improved.

A 16th aspect of the invention is directed to the light-emitting device as set forth in the 14th aspect of the invention, wherein the charge transport material has a substantially successive distribution of 25 concentration from the cathode side of the emission region toward the anode

side thereof.

With this constitution, the recombination efficiency of the electron and the hole is further improved and thus the luminance efficiency is improved.

A 17th aspect of the invention is directed to the light-emitting device  
5 as set forth in the 11th aspect of the invention, wherein the emission region includes a region where the material contributable to the emission is not present.

An 18th aspect of the invention is directed to the light-emitting device  
as set forth in the 11th aspect of the invention, wherein the medium for  
10 containing the material contributable to the emission has charge transport capability.

A 19th aspect of the invention is directed to the light-emitting device  
as set forth in the 11th aspect of the invention, wherein the medium for  
containing the material contributable to the emission comprises organic  
15 material.

A 20th aspect of the invention is directed to the light-emitting device  
as set forth in the 11th aspect of the invention, wherein the medium  
comprises polymer.

With this constitution in which the emission region comprises organic  
20 material, or to be more specific, polymer, the organic light-emitting device  
having an improved efficiency is produced.

A 21st aspect of the invention is directed to a display using the  
light-emitting device as set forth in the 11th aspect of the invention.

With this constitution, the display having an improved luminous  
25 efficiency is provided.

A 22nd aspect of the invention is directed to a producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising: a disposing step of disposing a medium on the anode or the cathode; and a containing step of allowing material contributable to emission to be contained in the medium to form the emission region.

Since this method comprises the step of disposing the medium on the anode or the cathode and the step of allowing material contributable to emission to be contained in the medium, it can provide an effective method when it is difficult to dispose the medium previously containing the material contributable to the emission on the anode or the cathode.

A 23rd aspect of the invention is directed to a producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising a disposing step of disposing a medium containing charge transport material on the anode or the cathode; and a containing step of allowing material contributable to emission to be contained in the medium to form the emission region.

This method enables the charge transport material to be contained in the medium, and as such can produce the light-emitting device that can inject and transport the charge with efficiency.

A 24th aspect of the invention is directed to a producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising a disposing step of disposing a medium on the anode or the cathode; and a containing step of allowing material contributable to emission and charge transport material to be

contained in the medium.

A 25th aspect of the invention is directed to a producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising a disposing step of disposing a medium containing charge transport material on the anode or the cathode; and a containing step of allowing material contributable to emission and charge transport material to be contained in the medium.

The charge of the charge transport material contained in the medium in the disposing step and the charge of the charge transport material in the 10 containing step may be different from or identical to each other.

A 26th aspect of the invention is directed to a producing method of a light-emitting device having a charge transport region between an anode and a cathode, the producing method comprising a disposing step of disposing a medium on the anode or the cathode; and a containing step of 15 allowing charge transport material to be contained in the medium.

A 27th aspect of the invention is directed to the producing method of a light-emitting device as set forth in the 23rd aspect of the invention, wherein in the containing step, the material contributable to the emission is penetrated into the medium, whereby it is contained in the medium.

20 A 28th aspect of the invention is directed to the producing method of a light-emitting device as set forth in the 24th aspect of the invention, wherein in the containing step, the material contributable to the emission and the charge transport material are penetrated into the medium, whereby they are contained in the medium.

25 This method provides the light-emitting device that enables the

material contributable to the emission, or the material contributable to the emission and the charge transport material to have a substantially successive distribution of concentration from the anode side of the charge transport region toward the cathode side thereof and vice versa.

5       A 29th aspect of the invention is directed to the producing method of a light-emitting device as set forth in the 23rd aspect of the invention, wherein in the containing step, solution obtained by the material contributable to the emission being dissolved in solvent is brought into contact with the medium, whereby the material is penetrated into the  
10      medium.

A 30th aspect of the invention is directed to the producing method of a light-emitting device as set forth in the 24th aspect of the invention, wherein in the containing step, solution obtained by the material contributable to the emission and the charge transport material being dissolved in solvent is brought into contact with the medium, whereby the materials are penetrated into the medium.  
15

This method also provides the light-emitting device that enables the material contributable to the emission, or the material contributable to the emission and the charge transport material to have a substantially successive distribution of concentration from the anode side of the charge  
20      transport region toward the cathode side thereof and vice versa.

A 31st aspect of the invention is directed to the producing method of a light-emitting device as set forth in the 27th aspect of the invention, wherein in the containing step, the material contributable to the emission is  
25      penetrated into the medium in an ink jet method.

The method above allows the material contributable to he emission to penetrate into the medium in the ink-jet method, so that when polymer high in viscosity is used as the precursor of the medium, the need of the application of the polymer solution and the material contributable to the 5 emission in the ink-jet method can be eliminated. Therefore, the nozzles of the ink jet are prevented from being plugged and also fine patterns can be formed with ease.

(Second inventive group)

On the basis of the concept that collection of the luminous molecules, 10 which were dispersed over the whole organic layer, to a specific region and enlargement of the surface area of the organic layer lead to improvement in efficiency of the polymer-based organic light-emitting device, the inventors of this application disperse the luminous molecules on a front surface of the organic layer or in a region in the vicinity of the front surface of the same 15 and thereby solve the problems mentioned above.

Specifically, a 32nd aspect of the invention is directed to a light-emitting device having an emission region between an anode and a cathode, wherein the emission region comprises material contributable to emission, and wherein a collecting means for collecting the material 20 contributable to the emission to a specific region is provided between the anode and the cathode.

To be more specific, a 33rd aspect of the invention is directed to a light-emitting device having an emission region between an anode and a cathode, wherein at least one of an anode side of the emission region and a 25 cathode side thereof is made porous, and wherein material contributable to

emission is included in a surface of the emission region which is made porous.

A 34th aspect of the invention is directed to a light-emitting device having an emission region between an anode and a cathode, wherein at 5 least one of an anode side of the emission region and a cathode side thereof is made porous, and wherein material contributable to emission is included in a region in the vicinity of a surface of the emission region which is made porous.

With the constitutions above, the material contributable to emission is 10 collected to a specific region, or specifically, to a surface of the emission region which is made porous or to a region in the vicinity of the surface thereof and also the recombination region of the hole and the electron in which the material contributable to emission is present is enlarged. Thus, the emission with improved luminance can be produced.

15 A 35th aspect of the invention is directed to the light-emitting device as set forth in the 33rd aspect of the invention, wherein charge transport material is included in a surface of the emission region which is made porous.

A 36th aspect of the invention is directed to the light-emitting device 20 as set forth in the 33rd aspect of the invention, wherein a leveled layer comprising charge transport material is provided on a surface of the emission region which is made porous.

With this constitution, leakage of current is prevented and also the injection and transport of the hole or the electron can be performed with 25 improved efficiency. Further, the leveled layer can permit the joint surface

of the adjacent anode and cathode to be kept smooth

A 37th aspect of the invention is directed to a light-emitting device having a charge transport region between an anode and a cathode, wherein at least one of an anode side of the charge transport region and a cathode side thereof is made porous.

With this constitution, the charge can be injected from the electrodes (anode and cathode) into the charge transport region with improved efficiency. In this constitution, the charge transport region is a region having the luminescent property and thus doubles as the property of the emission region.

A 38th aspect of the invention is directed to the light-emitting device as set forth in the 37th aspect of the invention, wherein the charge transport region is a hole transport region.

A 39th aspect of the invention is directed to the light-emitting device as set forth in the 37th aspect of the invention, wherein the charge transport region is an electron transport region.

A 40th aspect of the invention is directed to the light-emitting device as set forth in the 33rd aspect of the invention, wherein the emission region comprises an organic matter.

A 41st aspect of the invention is directed to the light-emitting device as set forth in the 33rd aspect of the invention, wherein the emission region comprises polymer.

A 42nd aspect of the invention is directed to a light-emitting device having an emission region between an anode and a cathode, wherein at least one of an anode side of the emission region and a cathode side thereof

is roughened, and wherein material contributable to emission is included in a surface of the roughened emission region.

A 43rd aspect of the invention is directed to a light-emitting device having an emission region between an anode and a cathode, wherein at 5 least one of an anode side of the emission region and a cathode side thereof is roughened, and wherein material contributable to emission is included in a region in the vicinity of a roughened surface of the emission region.

With these constitutions, the surface area of the recombination region of the hole and the electron where the material contributable to emission is 10 present is enlarged, thus producing the emission with improved luminance.

A 44th aspect of the invention is directed to the light-emitting device as set forth in the 42nd aspect of the invention, wherein a leveled layer comprising charge transport material is provided on a roughened surface of the emission region.

15 A 45th aspect of the invention is directed to a light-emitting device having a charge transport region between an anode and a cathode, wherein at least one of an anode side of the charge transport region and a cathode side thereof is roughened.

With this constitution, the charge can be injected from the electrodes 20 (anode and cathode) into the charge transport region with improved efficiency. When the emission region is located over the roughened charge transport region, the contact area of the emission region and the charge transport region is enhanced, so that the hole can be injected from the anode into the emission region with improved efficiency.

25 A 46th aspect of the invention is directed to the light-emitting device

as set forth in the 45th aspect of the invention, wherein the charge transport region is a hole transport region.

A 47th aspect of the invention is directed to the light-emitting device as set forth in the 45th aspect of the invention, wherein the charge 5 transport region is an electron transport region.

A 48th aspect of the invention is directed to the light-emitting device as set forth in the 42nd aspect of the invention, wherein the emission region comprises an organic matter.

A 49th aspect of the invention is directed to the light-emitting device 10 as set forth in the 42nd aspect of the invention, wherein the emission region comprises polymer.

A 50th aspect of the invention is directed to a display using the light-emitting device as set forth in the 33rd aspect of the invention.

A 51st aspect of the invention is directed to a lighting device using the 15 light-emitting device as set forth in the 33rd aspect of the invention.

With the constitutions above, the display and the lighting device having improved luminous efficiency can be provided.

A 52nd aspect of the invention is directed to a producing method of a light-emitting device having an emission region between an anode and a 20 cathode, the producing method comprising a medium disposing step of disposing a medium on the anode or the cathode; and a porosity producing step of making at least a part of the medium porous.

A 53rd aspect of the invention is directed to a producing method of a light-emitting device having an emission region between an anode and a 25 cathode, the producing method comprising a medium disposing step of

disposing a medium on the anode or the cathode; a porosity producing step of making at least one of an anode side of the medium and a cathode side thereof porous; and a disposing step of disposing material contributable to emission on a porous surface of the medium, so that the emission region is  
5 formed by the medium and the material contributable to the emission.

A 54th aspect of the invention is directed to a producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising a medium disposing step of disposing a medium on the anode or the cathode; a porosity producing step of making at least one of an anode side of the medium and a cathode side thereof porous; a containing step of allowing material contributable to emission to be contained in a region in the vicinity of a porous surface of the medium, so that the emission region is formed by the medium and the material contributable to the emission; and a disposing step of disposing  
10 charge transport material on a porous surface of the medium.  
15

With this constitution, the material contributable to the emission is collected to the porous surface of the emission region or the region in the vicinity of the porous surface of the same and also the surface area of the recombination region of the hole and the electron is enlarged, thus providing  
20 the light-emitting device with improved luminance.

A 55th aspect of the invention is directed to the producing method of a light-emitting device as set forth in the 53rd aspect of the invention, which comprises a disposing step of disposing charge transport material on a porous surface of the emission region.

25 A 56th aspect of the invention is directed to the producing method of a

light-emitting device as set forth in the 53rd aspect of the invention, which comprises a leveled layer forming step of forming a leveled layer comprising charge transport material on the emission region.

With the constitutions above in which the leveled layer is formed over the emission region, the light-emitting device is provided having the capabilities that leakage of current is prevented and also the injection and transport of the hole or the electron can be performed with improved efficiency and further, the leveled layer can permit the joint surface of the adjacent anode and cathode to be kept smooth.

10        A 57th aspect of the invention is directed to the producing method of a light-emitting device as set forth in the 53rd aspect of the invention, wherein the disposing step is a step of disposing a medium containing material soluble in a specified solvent, and the porosity producing step is a step of eluting the material from the solvent to thereby make the medium  
15        porous.

A 58th aspect of the invention is directed to a producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising a medium disposing step of disposing a medium on the anode or the cathode; and a roughening step of  
20        roughening a part of the medium.

A 59th aspect of the invention is directed to a producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising a medium disposing step of disposing a medium on the anode or the cathode; a roughening step of  
25        roughening at least one of an anode side of the medium and a cathode side

thereof; and a disposing step of disposing material contributable to emission on a roughened surface of the medium, so that the emission region is formed by the medium and the material contributable to the emission.

A 60th aspect of the invention is directed to a producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising a medium disposing step of disposing a medium on the anode or the cathode; a roughening step of roughening at least one of an anode side of the medium and a cathode side thereof; and a containing step of allowing material contributable to emission to be contained in a region in the vicinity of a roughened surface of the medium, so that the emission region is formed by the medium and the material contributable to the emission.

With the constitutions above, the material contributable to the emission is collected to the roughened surface of the emission region or the region in the vicinity of the roughened surface of the same and also the surface area of the recombination region of the hole and the electron where the material contributable to the emission is present is enlarged, thus providing the light-emitting device having the capability of producing improved luminance.

A 61st aspect of the invention is directed to the producing method of a light-emitting device as set forth in the 59th aspect of the invention, which comprises a leveled layer forming step of forming a leveled layer comprising charge transport material on the emission region.

With this constitution, the light-emitting device is provided having the capabilities that leakage of current is prevented and also the injection and

transport of the hole or the electron can be performed with improved efficiency and further, the joint surface of the adjacent anode and cathode can be kept smooth.

A 62nd aspect of the invention is directed to the producing method of a 5 light-emitting device as set forth in the 59th aspect of the invention, wherein the roughening step is a step of roughening the emission region by dry etching.

The dry etching enables the emission region to be roughened with ease.

10

#### Brief Description of the Drawings

FIG. 1 is a schematic sectional view of an organic light-emitting device according to an embodiment 1-1 of the present invention;

FIG. 2 shows schematic diagrams illustrating construction of an 15 organic light-emitting device according to an embodiment 1-2 of the present invention, FIG. 2(a) is a schematic conceptual diagram of the organic light-emitting device and FIG. 2(b) is a sectional view taken along the arrowed line A-A of FIG. 2(a);

FIG. 3 shows schematic sectional views illustrating the producing 20 method of the organic light-emitting device according to the embodiment 1-2 of the present invention;

FIG. 4 shows schematic sectional views illustrating another example of the producing method of the organic light-emitting device according to the embodiment 1-2 of the present invention;

25 FIG. 5 is a schematic diagram illustrating a distribution of

concentration of an emission region of the organic light-emitting device according to the embodiment 1·2 of the present invention;

FIG. 6 is a schematic sectional view of an organic light-emitting device according to an embodiment 1·3 of the present invention;

5 FIG. 7 is a schematic sectional view of an organic light-emitting device according to an embodiment 1·4 of the present invention;

FIG. 8 is a schematic sectional view of an organic light-emitting device according to an embodiment 1·5 of the present invention;

10 FIG. 9 is a schematic sectional view of another example of the organic light-emitting device according to the embodiment 1·5 of the present invention;

FIG. 10 is a schematic sectional view of yet another example of the organic light-emitting device according to the embodiment 1·5 of the present invention;

15 FIG. 11 is an energy level diagram of the organic light-emitting device according to the embodiment 1·5 of the present invention;

FIG. 12 is an energy level diagram of the organic light-emitting device according to the embodiment 1·5 of the present invention;

20 FIG. 13 is an energy level diagram of the organic light-emitting device according to the embodiment 1·5 of the present invention;

FIG. 14 is an energy level diagram of the organic light-emitting device according to the embodiment 1·5 of the present invention;

FIG. 15 is a schematic sectional view of an organic light-emitting device according to an embodiment 2·1 of the present invention;

25 FIG. 16 shows schematic sectional views illustrating the producing

method of the organic light-emitting device according to the embodiment 2-1 of the present invention;

FIG. 17 is a schematic sectional view showing a variant of the organic light-emitting device according to the embodiment 2-1 of the present invention;  
5

FIG. 18 is a schematic sectional view of an organic light-emitting device according to the embodiment 2-2 of the present invention;

FIG. 19 shows schematic sectional views illustrating the producing method of the organic light-emitting device according to the embodiment 2-2  
10 of the present invention; and

FIG. 20 is a schematic sectional view showing an organic light-emitting device according to an embodiment 2-3 of the present invention.

#### **Best Mode for Carrying out the Invention**

15 (Embodiments of First Inventive Group)

In the following, the first inventive group of the present invention will be described with reference to the accompanying drawings.

While in the embodiments illustrated below (the same applies to the second inventive group), description is given on an organic light-emitting  
20 device, the concept of the present invention above is applicable not only to the organic light-emitting device but also to an inorganic light-emitting device wherein an emission region is formed, for example, by inorganic phosphor being dispersed in an organic binder.

#### [Embodiment 1-1]

25 FIG. 1 is a schematic sectional view of an organic light-emitting device

according to an embodiment 1-1 of the present invention.

As shown in FIG. 1, an organic light-emitting device 10 comprises an anode 2 formed on a substrate 1, a cathode 4 arranged in opposition to the anode 2, and an emission region 3 interposed between the anode 2 and the cathode 4.

The emission region 3 comprises polymer 3A, luminous molecules 3G of material contributable to the emission, and charge transport materials 3F. In FIG. 1, only one kind of luminous molecules is shown.

The luminous molecules 3G and the charge transport materials 3F are distributed in concentration in a layer thickness direction of the emission region 3 (from the anode 1 to the cathode 4). Specifically, the concentration of the luminous molecules 3G and the charge transport materials 3F are large at a side of the emission region 3 close to the cathode 4 (the upper side as viewed in the diagram) and are small at a side thereof close to the anode 2 (the lower side as viewed in the diagram). Preferably, the emission region 3 has a region in the layer thickness direction thereof where neither the luminous molecules 3G nor the charge transport materials 3F are provided. In other words, it is preferable that the emission region 3 has, at the side thereof close to the anode 2, a region where neither the luminous molecules nor the charge transport material exists but only the polymer 3A exists.

While it is described that the luminous molecules 3G and the charge transport materials 3F are distributed in concentration in the layer thickness direction of the emission region, modification may be made such that only the luminous molecules 3G are distributed in concentration in the

layer thickness direction of the emission region 3 and the charge transport materials 3F are uniformly distributed in the layer thickness direction thereof.

Another modification may also be made of the organic light-emitting device, such that the emission region 3 comprises only the polymer 3A and the luminous molecules 3G (not include the charge transport materials 3F) and the luminous molecules 3G are distributed in concentration in the layer thickness direction.

An emission mechanism of the organic light-emitting device of the present invention is as follows. In the organic light-emitting device shown in FIG. 1, when a positive voltage is applied to the anode 2 and a negative voltage is applied to the cathode 4, the hole is injected into the emission region 3 from the anode 2 and the electron is injected into the emission region 3 from the cathode 4. Then, the injected hole flows to the cathode 4 and the injected electron flows to the anode 2. The hole and the electron are recombined in the emission region 3, in response to which fluorescence or phosphorescence is emitted from the luminous molecules 3G in the emission region 3.

The following points can be cited as major factors for determining current efficiency of the emission (emission efficiency for the injected current) include:

- (1) Efficiency of recombination of the hole and electron for the injected current;
- (2) Efficiency of generation of the excitons of the luminous molecules sequent from the recombination; and

(3) Efficiency of generation of luminous quantum from the excitons of the luminous molecules;

Of the factors above, (2) and (3) are practically determined by the luminous molecules' properties themselves.

5 On the other hand, the efficiency of recombination of the hole and electron of (1) is most influenced by the balance between the hole and the electron. If the hole and the electron are badly balanced, even if they are injected from the electrode, excess carriers reach the opposite electrode without being recombined in the emission region, resulting in wasteful  
10 currents that do not contribute to the emission.

If the mobility of the carriers in the emission region is enhanced, then the hole and the electron flow in a balanced manner and the luminous efficiency is improved. Specifically, it is preferable that the mobility of the hole is  $1 \times 10^{-7} \text{cm}^2/\text{V} \cdot \text{s}$  or more and the mobility of the electron is  $5 \times$   
15  $10^{-8} \text{cm}^2/\text{V} \cdot \text{s}$  or more.

When viewing from the emission mechanism mentioned above, in the case where the emission region includes a region of high hole transportability and a region of high electron transportability with respect to the layer thickness direction, or specifically, in the case where the hole  
20 transportability is high on the anode side of the emission region and the electron transportability is high on the cathode side of the same, the hole and the electron injected from the anode and the cathode respectively are locally recombined in the vicinity of the interface of the both regions.

Thus, when the luminous molecules 3G and the charge transport  
25 materials 3F are distributed in concentration in the layer thickness

direction, as shown in FIG. 1, or preferably, when the emission region 3 includes a region in which neither the luminous molecules 3G nor the charge transport materials 3F exists, a region different in carrier transportability is formed in the emission region 3, so that the recombination efficiency of the above-noted (1) is further improved and the emission efficiency is also improved.

[Embodiment 1-2]

FIG. 2 shows schematic diagrams illustrating construction of an organic light-emitting device according to an embodiment 1-2 of the present invention. FIG. 2(a) is a schematic conceptual diagram of the organic light-emitting device and FIG. 2(b) is a sectional view taken along the arrowed line A-A of FIG. 2(a).

As shown in FIG. 2, an organic light-emitting device 20 is a simple matrix type one comprising anodes 22 formed on a substrate 21 in a striped form, an emission region 23 formed on the anodes 22, and cathodes 24 formed on the emission region 23 in a striped form to extend orthogonal to the anodes 22.

The substrate 21 may be made of any material capable of supporting the organic light-emitting device 20 of the present invention. A transparent substrate made of glass or resin film such as polycarbonate, polymethyl methacrylate, and polyethylene terephthalate, or an opaque substrate made of silicon can be used as the substrate.

At least either of the anodes 22 and the cathodes 24 must be transparent or translucent to take out the light generated from the emission region 23 through the either or both of the electrodes.

While the transparent electrodes made of for example indium tin oxide (ITO) or tin oxide are often used as the anodes 22 in general, metallic electrodes made of, for example, Ni, Au, Pt, Pd, may be used. As for the ITO film, the deposition processes, such as a sputtering, an electron beam evaporation, an ion plating, is adopted, for the sake of improving the transparency or reducing the resistivity. The layer thickness is determined from a required sheet resistance and a required visible light transmittance, but since the organic light-emitting device is relatively large in drive current density, the film is often used with the thickness of 1,000 or more,  
5 in order to reduce the sheet resistance.  
10

Laminated electrodes of metal, such as Al, Ag and Au, alloy composed of metal of low in work function and metal of relatively large in work function and stable, such as MgAg alloy and AlLi alloy, and alloy composed of metal of low in work function and metal of high in work function, such as  
15 Li/Al and LiF/Al, can be used as the cathodes 24. These cathodes are preferably formed in the evaporation or the sputtering.

The anodes 22 and the cathodes 24 are the electrodes extending orthogonal to each other in the striped form. When a forward voltage is applied to selected anodes and cathodes, the emission region emits light  
20 with luminance corresponding to the applied voltage at the intersection points of the both of the electrodes.

While in this embodiment, the substrate 21, the anodes 22, the emission region 23 and the cathodes 24 are laminated in order from the bottom, they are not necessarily required to be laminated in this order.  
25 The order of lamination of these, from bottom to top, may be the substrate

21, the cathodes 24, the emission region 23 and the anodes 22.

If only the electrodes on the substrate 21 side or the anodes 22 are transparent and the cathodes 24 are opaque, then the substrate 21 is required to be transparent in order to take out the light generated.

5 Next, description on the emission region 23 will be given. The emission region 23 comprises polymer 23A, luminous molecules (Red) 23R, luminous molecules (Green) 23G and luminous molecules (Blue) 23B. The polymer 23A is seriated in an in-plane direction of the emission region 23, and a number of luminophors 23R, 23G, 23B are adjacently arranged in the  
10 direction parallel to the cathodes 24 and the anodes 22. The luminophors 23R, 23G, 23B are so distributed in density as to substantially continuously reduce from around the center of the emission region to the periphery in the direction parallel to the cathodes 24 and the anodes 22.

Now, description on the producing method of the organic light-emitting  
15 device according to the embodiment 1-2 of the present invention will be given. FIG. 3 shows schematic sectional views illustrating the producing method of the organic light-emitting device according to the embodiment 1-2 of the present invention.

(1) First, the anodes 22 made of ITO or equivalent are formed on the  
20 substrate 21 by the deposition method such as the sputtering, the electron beam evaporation, and the ion plating, as shown in FIG. 3(a).

(2) Then, the anodes 22 are patterned into a desired pattern (the striped form in this case) as shown in FIG. 3(b). For example, in the case of the ITO electrode, photo resist used is patterned by use of a common photo  
25 lithography and then is etched by hydriodic acid or equivalent.

- (3) Then, the polymer 23A of e.g. poly-N-vinylcarbazol is deposited on the anodes 22, as shown in FIG. 3(c). Though the deposition may be performed by any of the methods, such as the evaporation, the sputtering, and the method of application, it is performed mainly by the method of application. To be concrete, the solution of poly-N-vinylcarbazol dissolved in solvent such as toluene or chloroform is applied to the substrate 1 by the spin coat method or equivalent. The layer thickness of the polymer 3A is preferably in the range of about 500 to about 3,000 , though it is not limited to any particular thickness.
- 10 (4) Then, after the polymer 23A is deposited on the anodes, the luminous molecules having a desired luminous color is made to penetrate into a desired location, as shown in FIG. 3(d). Specifically, when a RGB full color panel is produced, the solution of the red luminous molecules 23R dissolved in the solvent is first discharged to drop on the red-color electrodes 15 (every two electrodes) of the striped anodes 22 formed of ITO in the ink-jet method using the ink heads 27, so as to penetrate into them.

- (5) Then, the solution of the green luminous molecules 23G is discharged onto the striped green-color anodes 22 by use of the ink heads 27 in the same manner, as shown in FIG. 3(e).
- 20 (6) Then, the solution of the blue luminous molecules 23B is discharged onto the blue-color anodes 22 in the same manner, as shown in FIG. 3(f). It is to be noted that any particular limitation is not imposed on the dropping order of the red, green and blue luminous molecules, and the luminous molecules may be dropped in any selected order. The luminous 25 molecules are preferably heat-treated, after dropping. The heat treatment

can accelerate the penetration of the luminous molecules 23R, 23G, 23B into the polymer 23A. For further acceleration of the penetration of the luminous molecules into the polymer 23A, it is preferable that a polymer-23A-soluble liquid is used as the solvent to dissolve the luminous  
5 molecules.

(7) Then, the striped cathodes 24 are formed to extend orthogonal to the striped anodes 22, as shown in FIG. 3(g). The deposition is performed in the evaporation method or the sputtering using the evaporation mask having a desired pattern.

10 While in the embodiment 1-2, the luminous molecules are penetrated into the polymer, the charge transport material as well as the luminous molecules may be penetrated into the polymer. Alternatively, the luminous molecules may be penetrated into the polymer in which the charge transport material is dispersed.

15 The method above allows the luminous molecules 23R, 23G and 23B to penetrate into the polymer in the ink-jet method, so that the need of the application of the polymer solution of high viscosity in the ink-jet method can be eliminated. Therefore, the luminous molecules can be applied without the nozzles of the ink jet being plugged, thus enabling the molecules  
20 to be formed into fine patterns.

Referring now to FIG. 5, description will be given on the configuration of the emission region of the organic light-emitting device produced in the producing method mentioned above. FIG. 5 is a schematic diagram illustrating a distribution of concentration of an emission region of the  
25 organic light-emitting device according to the embodiment 1-2 of the present

invention.

The luminous molecules (Red) 23R, the luminous molecules (Green) 23G and the luminous molecules (Blue) 23B are dispersed over the striped anodes 22 in the in-plane direction of the emission region 23 (in the 5 direction parallel to the electrodes), and the distribution of concentration of each luminous molecule 23R, 23G and 23B becomes the maximum at a substantially center point over the respective anode 22 and gradually reduces toward the both ends of the each anode 22, as shown in FIG. 5.

This constitution enables the luminous molecules 23R, 23G, 23B in the 10 vicinity of their boundaries to be small in concentration. Therefore, the luminous molecules 23R, 23G, 23B are prevented from being mixed in, thus providing little color mixture and thus producing an excellent full color display performance.

Further, the luminous molecules 23R, 23G, 23B are penetrated into 15 the polymer 23A in the ink-jet method using the ink-jets 27, so that the concentration of the luminance molecules 23R, 23G, 23B in the emission region 23 becomes larger on the cathode 24 side than on the anode 22 side.

Consequently, the organic light-emitting device of the embodiment 1-2 can provide increased recombination efficiency of the electron and the hole, 20 to produce improved luminous efficiency, as is the case with the embodiment 1-1 above.

FIG. 4 shows schematic sectional views illustrating another example of the producing method of the organic light-emitting device according to the embodiment 1-2 of the present invention.

25 The procedures shown up to FIG. 4(c) are the same as those of FIG. 3

that the polymer 3A is formed over the substrate 1 on which the anode 2 is formed.

Then, the luminous molecules 3R, 3G, 3B are penetrated in the polymer 3A in the following manner.

5       First, after a mask 8 having opening formed only on the red-color electrode of the striped anodes 2 of ITO is set on the substrate 1, steaming is performed by use of solution of red luminous molecules dissolved in the solvent, as shown in FIG. 4(d), so that the red luminous molecules 3R are penetrated into the polymer at the desired locations.

10      Sequentially, the green luminous molecules 3G and the blue luminous molecules 3B are penetrated in the same manner, as shown in FIG. 4(e),(f). It is to be noted that any particular limitation is not imposed on the steaming order of the red, green and blue luminous molecules, and the luminous molecules may be steamed in any selected order. Preferably, the 15 luminous molecules are heat-treated after steamed. The heat-treatment can accelerate the penetration of the luminous molecules 3B-3D into the polymer 3A. For further acceleration of the penetration of the luminous molecules into the polymer 3A, it is preferable that the polymer-3A-soluble liquid is used as the solvent to dissolve the luminous molecules.

20      Finally, the striped cathodes 4 are formed to extend orthogonal to the striped anodes 2 in the same manner as in FIG. 3.

While in the embodiment above, the luminous molecules are penetrated into the polymer, the charge transport material as well as the luminous molecules may be penetrated into the polymer. Alternatively, the 25 luminous molecules may be penetrated into the polymer in which the charge

transport material is dispersed.

Also, the method of penetrating the luminous molecules into the polymer in the printing method can be cited as another example of the producing method of the organic light-emitting device according to the  
5 present invention.

Specifically, in place of the luminous molecules 3R, 3G, 3B being penetrated into the polymer 3A in the steaming in FIG. 4(d)-(f), solution of the luminous molecules is applied to and penetrated into the polymer 3A by use of an offset printing method or a screen printing method.

10 In this variant as well, the luminous molecules are preferably heat-treated after printed, as is the case with the example noted above. Further, the polymer 3A-soluble liquid is used as the solvent to dissolve the luminous molecules. Instead of the luminous molecules being penetrated into the polymer, the charge transport material as well as the luminous  
15 molecules may be penetrated into the polymer. Alternatively, the luminous molecules may be penetrated into the polymer in which the charge transport material is dispersed.

#### [Embodiment 1-3]

FIG. 6 is a schematic sectional view of an organic light-emitting device  
20 according to an embodiment 1-3 of the present invention.

The embodiment 1-3 differs from the embodiment 1-2 in that in addition to the luminous molecules 23R, 23G, 23B, the charge transport material 23E are presented in the emission region 23. Here, the polymer 23A and the charge transport material 23E are seriated in the in-plane  
25 direction of the emission region 23. In other words, the charge transport

material 23E is uniformly dispersed in the polymer 23A. The luminous molecules 23R, 23G, 23B are dispersed over the striped anodes 22, respectively, as is the case with the embodiment 1-2.

[Embodiment 1-4]

5 FIG. 7 is a schematic sectional view of an organic light-emitting device according to an embodiment 1-4 of the present invention.

FIG. 7 differs from FIG. 6 in that the charge transport material 23E are not seriated in the in-plane direction of the emission region 23, but are distributed in concentration in the in-plane direction, as is the case with the 10 luminous molecules 23R, 23G and 23B. Specifically, the charge transport material 23E are present in large concentration in a region where the luminous molecules 23R, 23G, 23B are large in concentration.

[Commonalities among Embodiments 1-2 to 1-4]

Polymer having charge transport capabilities (charge transport 15 polymer) is preferably used as the polymeric material. Among others, polymer having hole transport capabilities (hole transport polymer) is preferable. Preferably, the hole transport polymer has the mobility of carrier of  $1 \times 10^{-7} \text{cm}^2/\text{V} \cdot \text{s}$  or more. Particularly preferable is poly-N-vinylcarbazol.

20 When the hole transport polymer is used, the electron transport material is preferably used as the charge transport material. Further, it is preferable that the electron transport material has the mobility of the carrier of  $5 \times 10^{-8} \text{cm}^2/\text{V} \cdot \text{s}$  or more. Particularly preferable are oxazole derivative, oxadiazole derivative, triazole derivative, pyrazine derivative, 25 aldazine derivative, quinolinol complex and derivative thereof. A content of

the electron transport material to the polymer is preferably 30-120 weight%. With the electron transport material content of less than 30 weight%, the electron transportability is not sufficient, while on the other hand, with the electron transport material content of more than 120 weight%, the dispersibility into the polymer deteriorates.

Fluophor or phosphor that exhibits luminescence in response to the recombination of the hole and electron may be used as the luminous molecules. The materials that emit particularly strong fluonescence or phosphorescence, which may be used, include coloring matters or laser dyes, such as cyanine dye, merocyanine dye, styrylic dye, anthracene derivative, porphyrin derivative, phthalocyanine derivative, coumarin, DCM and Nile red. Preferably used as the luminous molecule is the material having ionization potential of the luminous molecule smaller than ionization potential of the hole transport polymer and also electron affinity of the luminous molecule larger than electron affinity of the electron transport material.

[Embodiment 1-5]

FIGS. 8-10 are schematic sectional views of the organic light-emitting device according to an embodiment 1-5 of the present invention. In FIGS. 20 8-10, 25 designates a hole injection layer, 26 designates an electron injection layer, 23G designates the luminous molecules, 23H designates hole transport material, and 23I designates electron transport material.

The hole injection layer 25 is inserted for the purpose of subserving the injection of the hole from the anode 22 into the emission region 23. 25 Preferably, the material used for the hole injection layer 25 has the relation

among the ionization potential of the hole injection layer ( $Ip(h)$ ), the ionization potential of the polymer ( $Ip(p)$ ) and the ionization potential of the anode or the work function ( $Ip(a)$ ) of the anode being  $Ip(a) < Ip(h) < Ip(p)$ . It is particularly preferable that the hole injection layer comprises at least one material of polyaniline derivative, polythiophene derivative and amorphous carbon.

The electron injection layer 26 is inserted for the purpose of subserving the injection of the electron from the cathode 24 into the emission region 23. Desirably, the material used for the electron injection layer 26 has the electron affinity or work function smaller than the work function of the cathode. It is particularly preferable that the electron injection layer comprises at least one material of dilithium phthalocyanine, disodium phthalocyanine and organic boron complex compound.

The hole transport material 23H is introduced for the purpose of subserving the injection of the hole from the anode 22 into the emission region 23, as is the case with the hole injection layer. However, differently from the hole injection layer 25, the hole transport material is not inserted in between the anode 22 and the emission region 23 in the form of a layer, but is directly dispersed in the emission region.

Preferably, the material used for the hole transport material 23H has the ionization potential smaller than the ionization potential of the polymer. A content of the hole transport material to the polymer is preferably 10-120 weight%. With the hole transport material content of less than 10 weight%, the hole cannot be injected fully, while on the other hand, with the hole transport material content of more than 120 weight%, the dispersibility into

the polymer deteriorates.

The effects produced by the introduction of the hole injection layer 25, the electron injection layer 26 and the hole transport material 23H will be described with reference to the related diagrams. FIGS. 11-14 are energy level diagrams of the organic light-emitting device according to the present invention.

FIG. 11 shows an energy level diagram and an operation mechanism of an organic light-emitting device having the structure of anode/emission region (hole transport polymer + electron transport material + luminous molecules (luminescent material))/cathode. FIG. 12 shows an energy level diagram and an operation mechanism of an organic light-emitting device having the structure of anode/hole injection layer/emission region (hole transport polymer + electron transport material + luminous molecule/cathode. As already noted, when a voltage is applied to the organic light-emitting device, the hole is injected from the anode into the emission region and the electron is injected from the cathode into the emission region. In more detail, as shown in FIG. 11, the both carriers are injected into the material of smaller injection barrier. In other words, the hole is injected into the hole transport polymer in the emission region, and the electron is injected into the electron transport material in the emission region. It should be noted that the smaller the injection barrier for the both carriers (hole and electron) become, the more the injection of the carriers facilitates and the more the driving voltage reduces. This reduction of the driving voltage can provide improved power efficiency for emission (emitting efficiency for input power) even when the current

efficiency is identical. For example, when the hole injection layer having the ionization potential between the anode and the hole transport polymer is inserted, the hole injection barrier is relaxed, as shown in FIG. 12, and the driving voltage can also be lowered. Further, when the hole injection  
5 barrier is greater than the electron injection barrier, as shown in FIG. 11, the hole injection barrier is lowered so that an improved balance between the injection rate of the hole and the injection rate of the electron is produced, and as such can allow the current efficiency to be expectably improved by the aforesaid effects. FIG. 13 shows an energy level diagram  
10 of an organic light-emitting device having the structure of anode/emission region (hole transport polymer + electron transport material + luminous molecules)/electron injection layer/cathode. As is the case with the hole injection barrier, the electron injection barrier can also be lowered by inserting the electron injection layer smaller in electron affinity than the  
15 cathode, as shown in FIG. 13, thus producing reduction in driving voltage and improvement in luminous efficiency. FIG. 14 shows an energy level diagram of an organic light-emitting device having the structure of anode/emission region (hole transport polymer + hole transport material + electron transport material + luminous molecules)/cathode. In this case,  
20 since the ionization potential of the hole transport material is smaller than that of the hole transport polymer, the hole is directly injected into the hole transport material in the emission region from the anode, as shown in the diagram, so that the injection barrier is lowered, as compared with the case of the hole being injected into the hole transport polymer. Hence, as is the  
25 case with the structure of the hole injection layer being inserted, the driving

voltage can be reduced and at the same time the current efficiency can be expectedly improved.

Of course, the above-noted structures may be combined to provide the structure wherein both of the hole injection layer and the electron injection  
5 layer are inserted; the structure wherein the emission region comprises the hole transport polymer + hole transport material + electron transport material + luminous molecules and further the electron injection layer is inserted; or the structure wherein the luminous molecules and the charge transport material are distributed in concentration in the layer thickness  
10 direction of the emission region.

While in the above-mentioned embodiments 1·1 to 1·5, examples of the simple matrix organic light-emitting device are shown, modification may be made, such as, for example, forming the light emitting device structured as mentioned above on the thin film transistor to produce an active matrix  
15 display panel.

Now, experimental examples based on the embodiments mentioned above will be described in further detail.

(Experimental Example 1)

The organic light-emitting device was produced as mentioned below, in  
20 accordance with the procedures of FIG. 3 illustrated in the aforesaid Embodiment 1·1.

A glass substrate having thickness of 0.7mm was used as the substrate  
1 and ITO was deposited thereon in the form of the anode 2 by the sputtering method. The ITO was deposited to have thickness of about  
25 1,000 and the sheet resistance was set at about  $150\Omega/\square$ . The ITO was

patterned in the striped form having width of  $300\mu\text{m}$  by use of photo lithography.

Then, after the substrate was washed and subjected to oxygen plasma treatment, poly-N-vinylcarbazol (PVK)(molecular weight of about 28,000) 5 was deposited thereon as the polymer 3A. The PVK is the hole transport polymer and the mobility of the carriers is about  $2\times 10^{-6}\text{cm}^2/\text{V}\cdot\text{s}$ . The deposition was performed in the spin coat method, using the solution in which 300mg of PVK was dissolved in 30ml of toluene. The spin coat was performed with spin in a closed state in the conditions of 500rpm/10sec. and 10 of 1,000rpm/30sec..

Then, the PVK was heat-treated at  $110^\circ\text{C}$  for 1 minute by use of a hot plate. It was about 1,000 in layer thickness.

Then, the luminous molecules were dropped on the ITO electrodes at desired locations by use of a commercially available ink-jet printer, to form 15 the emission region. Nile red was used as red luminous molecules (3R), coumarin 6 was used as green luminous molecules (3G), and coumarin 47 was used as blue luminous molecules (3B). The respective luminous molecules were discharged from the ink head 7 in the form of solution in which 1mg of luminous molecules were dissolved 10ml of chloroform. 20 Every time the luminous molecules were dropped, they were heat-treated at  $110^\circ\text{C}$  for 1 minute by use of the hot plate.

Finally, Li/Al laminated electrodes were deposited as the cathodes 4 by the vacuum evaporation method. The deposition was performed under the degree of vacuum of about  $5\times 10^{-6}$  Torr. First, Li was allowed to evaporate 25 by 10 at a rate of about 0.5 /sec. and then Al was allowed to evaporate by

1,500 at a rate of about 30 /sec.. The cathodes was formed into the striped form orthogonal to the anodes 2 by use of the evaporation mask and having the width of  $300\mu\text{m}$ .

The energy levels of the respective materials are as follows. The  
5 ionization potential of the ITO is 4.9eV, the ionization potential of the PVK  
is 5.6eV, the electron affinity is 2.0eV, the ionization potential of Nile red is  
5.3eV, the electron affinity is 3.5eV, the ionization potential of coumarin 6 is  
5.4eV, the electron affinity is 2.9eV, the ionization potential of coumarin 47  
is 5.4eV, and the electron affinity is 2.5eV. The work function of Li is 2.9eV  
10 and the work function of Al is 4.3eV.

When an elemental analysis of the region where the coumarin 6 was dropped was made with respect to the layer thickness direction, in order to examine the distribution of concentration of the luminous molecules in the emission region 3, it was found that the quantity of sulfur contained only in  
15 the coumarin 6 gradually decreased from the cathode side toward the anode side and there were no coumarin 6 left in the vicinity of the anode.

In the organic light-emitting device thus produced, when a forward voltage of the order of 10V was applied across the selected striped anode and cathode, the part (critical area) sandwiched between the both electrodes  
20 emitted light brightly in the respective colors corresponding to the luminous molecules (Nile red: red, coumarin 6: green, and coumarin 47: blue). Thus, the simple matrix display capable of emitting light in the desired colors at desired locations (critical areas) was produced.

The current efficiency of each luminescent color (cd/A), the driving  
25 voltage at a luminance of  $100\text{cd}/\text{m}^2$  and the power efficiency ( $\text{lm}/\text{W}$ ) at a

luminance of 100cd/m<sup>2</sup> are shown in TABLE 1.

TABLE 1

	Current efficiency (cd/A)			Driving voltage (at 100cd/m <sup>2</sup> )(V)			Power efficiency (at 100cd/m <sup>2</sup> )(lm/W)		
	Red	Green	Blue	Red	Green	Blue	Red	Green	Blue
Experimental Example 1	1.5	3.0	1.7	12.0	11.0	12.0	0.4	0.9	0.4
Experimental Example 2	3.0	6.0	3.4	10.4	9.0	10.0	0.9	2.1	1.1
Experimental Example 3	3.3	7.2	4.0	9.2	8.0	9.0	1.1	2.8	1.4
Experimental Example 4	1.8	4.1	2.0	10.5	9.2	10.3	0.5	1.4	0.6
Experimental Example 5	1.9	4.2	2.0	10.5	9.0	10.0	0.6	1.5	0.6
Experimental Example 6	1.9	4.2	2.1	10.4	9.0	10.1	0.6	1.5	0.7
Experimental Example 7	1.8	4.1	2.0	10.5	9.0	10.0	0.5	1.4	0.6
Experimental Example 8	1.7	4.0	2.1	10.2	8.8	9.9	0.5	1.4	0.7
Experimental Example 9	1.8	4.5	2.2	10.3	8.9	9.9	0.5	1.6	0.7
Experimental Example 10	3.5	7.1	3.8	9.0	7.6	8.7	1.2	2.9	1.4
Experimental Example 11	1.6	3.2	1.5	12.0	11.0	12.1	0.4	0.9	0.4
Experimental Example 12	3.0	6.1	3.2	10.4	9.0	10.1	0.9	2.1	1.0
Experimental Example 13	3.4	7.0	4.0	9.2	8.0	9.1	1.2	2.7	1.4
Experimental Example 14	1.5	3.1	1.6	10.5	9.2	10.2	0.4	1.1	0.5
Experimental Example 15	3.0	6.1	3.0	10.5	9.0	10.0	0.9	2.1	0.9
Experimental Example 16	3.5	7.2	3.9	10.4	9.0	10.1	1.1	2.5	1.2

5

(Experimental Example 2)

In Experimental example 1, in place of the PVK being deposited as the polymer 3A, the PVK in which 2-(4-biphenyl)-5-(4-tert butylphenyl)-1,3,4-oxadiazole (PBD) was dispersed. The PBD is the

electron transport material and the mobility of the carriers is about  $2 \times 10^{-6} \text{cm}^2/\text{V} \cdot \text{s}$ . The ionization potential is 6.1eV and the electron affinity is 2.4eV. The deposition was performed in the same conditions as those of the experimental example 1 by the spin coat method, using the solution in which 300mg of PVK and 180mg of PBD are dissolved in 30ml of solvent in which toluene and chloroform were mixed in the proportion 1:1.

5 The properties of this device are shown in TABLE 1.

(Experimental Example 3)

In Experimental example 1, in place of the luminous molecules being dropped, the mixed solution of the luminous molecules and the PBD (electron transport material) was dropped.

10 Specifically, the solution in which 1mg of Nile red + 100mg of PBD, 1mg of coumarin 6 + 100mg of PBD, and 1mg of coumarin 47 + 100mg of PBD were respectively dissolved in 30ml of solvent in which toluene and 15 chloroform were mixed in the proportion 1:1 was dropped on the PVK by the ink jet method.

The properties of this device are shown in TABLE 1.

(Experimental Example 4)

In Experimental example 1, the hole injection layer was inserted in 20 between the anode and the emission region.

In short, the device was structured, as shown in FIG. 8. The hole injection layer was formed by the spin coat method using a commercially available polythiophene derivative, to have layer thickness of 150 , before the deposition of the PVK. The ionization potential of the polythiophene 25 derivative used here is 5.3eV.

The properties of this device are shown in TABLE 1.

(Experimental Example 5)

As a substitution for the polythiophene derivative as the hole injection layer of the experimental example 4, a commercially available polyaniline derivative that exhibits equivalent ionization potential to the above-noted polythiophene derivative was used. The deposition of the polyaniline derivative was performed in the same manner as in the experimental example 2, to have layer thickness of 150 .

The properties of this device are shown in TABLE 1.

10 (Experimental Example 6)

As a substitution for the polythiophene derivative as the hole injection layer of the experimental example 4, amorphous carbon was used. The amorphous carbon was formed to have layer thickness of 100 by the sputtering method. The ionization potential of the amorphous carbon is  
15 5.2eV.

The properties of this device are shown in TABLE 1.

(Experimental Example 7)

In Experimental example 1, the electron injection layer was inserted in between the emission region and the cathode.

20 In short, the device was structured, as shown in FIG. 9. Dilithium phthalocyanine was used and deposited as the electron injection layer by the vacuum evaporation method after the luminous molecules were dropped over the PVK. Sequentially, Al was deposited as the cathode. The electron injection layer and the cathode were formed in the manner that  
25 after dilithium phthalocyanine was deposited 10 at a rate of about 0.3

/sec., Al was deposited 1,500 at a rate of about 30 /sec.. The electron affinity of dilithium phthalocyanine is 3.0eV.

The properties of this device are shown in TABLE 1.

(Experimental Example 8)

- 5 As a substitution for the dilithium phthalocyanine as the electron injection layer of the experimental example 7, disodium phthalocyanine that exhibits equivalent electron affinity thereto was used. The deposition of the disodium phthalocyanine was performed in the same manner as in the experimental example 7, to have layer thickness of 10 .
- 10 The properties of this device are shown in TABLE 1.

(Experimental Example 9)

- As a substitution for the dilithium phthalocyanine as the electron injection layer of the experimental example 7, 4,4,8,8-tetrakis (1H-pyrazole-1-yl) pyrazabole was used. The deposition of the 15 4,4,8,8-tetrakis (1H-pyrazole-1-yl) pyrazabole was performed in the same manner as in the experimental example 7, to have layer thickness of 10 . The electron affinity of the 4,4,8,8-tetrakis (1H-pyrazole-1-yl) pyrazabole is 2.3eV

The properties of this device are shown in TABLE 1.

- 20 (Experimental Example 10)

- In the experimental example 3, in place of PVK being deposited as the polymer 3A, the PVK in which N,N'-diphenyl-N,N'-bis (3-methylphenyl)-1,1'-biphenyl-4,4'-diamine (TPD) is dispersed as the hole transport material. In short, the device shown in FIG. 10 was produced.
- 25 The ionization potential of TPD is 5.4eV. The deposition was performed in

the same conditions as those of the experimental example 1 by the spin coat method, using the solution in which 300mg of PVK and 180mg of PBD were dissolved in 30ml of mixed solvent in which toluene and chloroform were mixed in the proportion 1:1.

5       The properties of this device are shown in TABLE 1.

(Experimental Example 11)

In accordance with the procedure of FIG. 4, the organic light-emitting device was produced. After the procedures until FIG. 4(c) were performed in the same manner as in the experimental example 1, the luminous 10 molecules are penetrated into the polymer 3A by steaming, to form the emission region. The luminous molecules used Nile red, coumarin 6 and coumarin 47, as is the case with the experimental example 1. The steaming was performed in the following manner. First, after the mask having the openings at portions corresponding to the red-color electrodes of 15 the striped anodes 2 formed of ITO was set on the substrate, the polymer was subjected to the steam generated by heating the solution in which 10mg of Nile red was dissolved in 10ml of toluene. Further, it was heat-treated at 110°C for 1 minute by use of a hot plate. Thus, the Nile red was penetrated into the substrate at the desired positions thereof. Sequentially, 20 after the mask was shifted in position so that the openings were over the green-color electrodes, the polymer was subjected to the steam generated by heating the solution in which 10mg of coumarin 6 was dissolved in 10ml of toluene and further was heat-treated at 110°C for 1 minute. Sequentially, after the mask was shifted in position in the same manner, the polymer was 25 treated by the steaming using the solution in which 10mg of coumarin 47

was dissolved in 10ml of toluene and further was heat-treated at 110°C for 1 minute. Finally, the Li/Al laminated electrodes were deposited as the cathodes 24 in the vacuum evaporation method in the same manner as in the experimental example 1.

5 The properties of this device are shown in TABLE 1.

(Experimental Example 12)

In the experimental example 11, in place of PVK being deposited as the polymer 3A, the PVK in which 2-(4-biphenyl)-5-(4-tert butylphenyl)-1,3,4-oxadiazole (PBD) was dispersed was deposited. The deposition was performed in the same conditions as those of the experimental example 1 by the spin coat method, using the solution in which 300mg of PVK and 180mg of PBD were dissolved in 30ml of mixed solvent in which toluene and chloroform were mixed in the proportion 1:1.

The properties of this device are shown in TABLE 1.

15 (Experimental Example 13)

In the experimental example 11, in place of the luminous molecules being steamed, the mixed solution of the luminous molecules and the PBD was steamed.

Specifically, the solution in which 1mg of Nile red + 100mg of PBD, 20 1mg of coumarin 6 + 100mg of PBD, and 1mg of coumarin 47 + 100mg of PBD were respectively dissolved in 30ml of mixed solvent in which toluene and chloroform were mixed in the proportion 1:1 was used and steamed on the PVK.

The properties of this device are shown in TABLE 1.

25 (Experimental Example 14)

In the experimental example 1, in place of the luminous molecules being dropped, the luminous molecules were penetrated in the printing method. After the PVK was deposited on the substrate in the same procedure as in the experimental example 1, the solution in which 1mg of  
5 Nile red was dissolved in 10mg of toluene was applied to the desired ITO electrodes in the screen printing method and then was heat-treated at 10°C for 1 minute by use of the hot plate. Likewise, the solution in which 1mg of coumarin 6 and coumarin 47 were dissolved in 10ml of toluene was applied to the desired ITO electrodes in the screen printing method and then was  
10 heat-treated at 110°C for 1 minute by use of the hot plate. Finally, the Li/Al laminated electrodes were deposited as the cathodes 24 in the vacuum evaporation method in the same manner as in the experimental example 1.

The properties of this device are shown in TABLE 1.

(Experimental Example 15)

15 In the experimental example 14, in place of PVK being deposited as the polymer 3A, the PVK in which 2-(4-biphenyl)-5-(4-tert butylphenyl)-1,3,4-oxadiazole (PBD) was dispersed was deposited. The deposition was performed in the same conditions as those of the experimental example 1 by the spin coat method, using the solution in  
20 which 300mg of PVK and 180mg of PBD were dissolved in 30ml of mixed solvent in which toluene and chloroform were mixed in the proportion 1:1.

The properties of this device are shown in TABLE 1.

(Experimental Example 16)

In the experimental example 14, in place of the luminous molecules  
25 being applied in the screen printing method, the mixed solution of the

luminous molecules and the PBD was applied in the screen printing method. Specifically, the solution in which 1mg of Nile red + 100mg of PBD, 1mg of coumarin 6 + 100mg of PBD, and 1mg of coumarin 47 + 100mg of PBD were respectively dissolved in 30ml of mixed solvent in which toluene and 5 chloroform were mixed in the proportion 1:1 was applied onto the PVK in the screen printing method.

The properties of this device are shown in TABLE 1.

(Embodiments of Second Inventive Group)

The light-emitting device of the second inventive group is 10 characterized in that it comprises the emission region provided between the anode and the cathode and comprising material contributable to the emission and also comprises a collecting means, provided between the anode and the cathode, for collecting the material contributable to the emission into a specific region. To be more concrete, the light-emitting device 15 comprises the emission region provided between the anode and the cathode and comprising material contributable to the emission; a porous or roughened region between the anode and the cathode; and material contributable to the emission which is arranged on as well as in the vicinity of a front surface of the porous or roughened region.

As mentioned above, the provision of the porous or roughened region 20 can allow the material contributable to the emission to be collected to a specific region (on or in the vicinity of the front surface of the porous or roughened region) and also can enlarge a surface area of the hole-and-electron recombination region where the material contributable to 25 the emission is present. This can produce the emission with high

luminance.

The application of the light-emitting device thus constructed to a display or a lighting device can produce the display or lighting device having high luminance performance.

5 In the following, the embodiment 2-1 illustrates the light-emitting device having the structure in which the emission region is made porous. The embodiment 2-2 illustrates the structure in which the emission region is roughened. Further, the embodiment 2-3 illustrates the structure in which the charge transport region is made porous or is roughened.

10 [Embodiment 2-1]

FIG. 15 is a schematic sectional view of an organic light-emitting device according to an embodiment 2-1 of the present invention.

As shown in FIG. 15, an organic light-emitting device 100 comprises an anode 105 formed on a substrate (not shown), a cathode 101 arranged in 15 opposition to the anode 105, an emission region 109 arranged between the anode 105 and the cathode 101, and a leveled layer 102 arranged between the cathode 101 and the emission region 109.

The emission region 109 comprises an organic layer 104 which is made porous on the cathode 101 side thereof, and luminance molecules 103 arranged on a front surface of the organic layer 104 thus made porous.

Further, a leveled layer 102 comprising charge transport material is formed on the front surface of the organic layer 104 and the cathode 101 is laminated on the leveled layer 102.

This constitution, in which the luminous molecules 103 are collected 25 on the front surface of the organic layer 104 as was made porous to enlarge

a surface area thereof and also the hole-and-electron recombination region in which the luminous molecules 103 are present is made porous to enlarge a surface area thereof, can produce the emission with increased luminance.

In addition, the leveled layer 102 comprising the charge transport material 5 is provided on the surface of the organic layer 104 as was made porous, thus producing the results that joint surfaces of adjoining electrodes (cathodes 101) can be kept smooth; that leakage of current can be prevented; and that the hole or the electron can be injected and transported with efficiency.

Also, the filling of the charge transport material into the porous 10 organic layer 104 enables the hole or the electron to be injected or transported with efficiency.

The coloring matters whose quantum efficiency is nearly 1, including laser dyes such as coumarin 6, DCM and phenoxazone 9, are preferably used for the luminous molecules 103. In addition to this, fused rings, such 15 as naphthalene, anthracene, pyrene, and naphthacene, and derivatives thereof are also preferable. For example, rubrene has quantum efficiency of nearly 1 and is also an efficient luminous material. In addition, metal complexes, such as Alq and derivative thereof and beryllium benzoquinoline, are also preferable.

20 The charge transport material that is filled in the porous organic layer 104 or forms the leveled layer 102 must transport the charge reverse in polarity to the charge transported by the porous organic layer 104.

When the porous organic layer 104 is formed as the hole transport 25 organic layer, the electron transport material is chosen as the charge transport material. On the other hand, when it is formed as the electron

transport organic layer, the hole transport material is chosen as the charge transport material.

The electron transport materials that may preferably be used include low molecular weight material easy to enter the interior of the porous layer.

5      Metal complex, such as Alq and tris (4-methyl-8-quinolinolato) aluminum, electron deficient compound, such as 4,4,8,8-tetrakis (1H-pyrazole-1-yl) pyrazabole, and 3-(2'-benzothiazolyl)-7-diethylaminocoumarin can be cited as the preferable material. Materials having the hole blocking function, such as bathocuproine and triazole derivative, are also preferable.

10     The hole transport materials that may be preferably be used include derivative having triphenylamine as fundamental skeleton. For example, tetraphenylbenzidine compound, triphenylamine trimer and benzidine dimer disclosed by Japanese Laid-open (unexamined) Patent Publication No. Hei 7-126615, various triphenyldiamine derivatives disclosed by Japanese Laid-open (unexamined) Patent Publication No. Hei 8-48656, and MTPD (common name: TPD) disclosed by Japanese Laid-open (unexamined) Patent Publication No. Hei 7-65958 are preferable.

The hole transport material, when filled in the porous layer, can be replaced by the electron injection material.

20     The electron injection materials that may preferably be used include dilithium phthalocyanine, disodium phthalocyanine, magnesium porphin, 4,4,8,8-tetrakis (1H-pyrazole-1-yl) pyrazabole disclosed by Japanese Patent Application No. Hei 11-214712. The hole injection material that may preferably used include copper phthalocyanine, 5,10,15,20-tetraphenyl-21H,23H-porphin copper (In Embodiment 2-2

mentioned later, even when the roughened surface is smoothened to the order of 10nm, the electron injection material may be replaced by the charge injection material).

In addition to the low molecular weight material, polymeric material 5 may be chosen as the material to be filled in the porous layer, but since there is the possibility that the material could not be fully filled therein by only the application, some measurement, such as heat-treatment at glass transition point, should be taken to thoroughly penetrate the material into the porous layer (In Embodiment 2-2 mentioned later, the polymeric 10 material may be chosen even for filling the roughened surface).

In consideration of making the organic layer porous, organic polymer is preferable for the material to form the organic layer 104. Particularly, when it is made to be porous, the polymer-based organic compound is chosen, since it is subjected to the wet etching process (In Embodiment 2-2 15 mentioned later, the organic polymer is preferable even for filling the roughened surface).

Material for general purpose applications, such as poly-p-phenylenevinylene (PPV), polyvinyl carbazol (PVK), polymethylmethacrylate (PMMA), polyfluorene and derivatives thereof can 20 be used as the organic polymers to form the organic layer. Further, for the purpose of improving the charge transport capabilities, the electron transport material and the hole transport material may be mixed. The material for general purpose applications may be used.

Preferably, the organic layer is formed to have thickness of 10·1,000nm. 25 Preferably, the thickness of the region to be made porous of the organic layer

falls within about one-third of the whole thickness of the organic layer, or particularly preferably in the range of 5-50nm. When the organic layer is reduced in thickness to 10nm or less, short-circuit is caused when a voltage is applied. On the other hand, when it is increased to 1,000nm or more, the  
5 applied voltage is increased and the luminous efficiency is reduced. Also, when the region to be made porous is more than one-third of the whole organic layer or more than 50nm, the organic layer is reduced in density, so that the charge transport performance is deteriorated and also stiffness and adhesion of the layer to the substrate is weakened. On the other hand,  
10 when the region to be made porous is less than 5nm, the above-mentioned effects are not easily produced (In the embodiment 2-2 mentioned later, the same applies to the region to be roughened). In the applied organic layer, it is necessary that the uniform layer should be formed by deposition, first, from the viewpoint of the injection and transport of charge. When viewed  
15 microscopically, the organic polymer layer is rough, but the term of "uniform layer" used herein means that the layer is uniform when viewed macroscopically. It is necessary that the roughness of the surface of the deposited layer should be set within 5nm at least. Making the organic polymer layer porous and rough means that the roughness of the uniformly  
20 deposited layer is further increased.

Referring now to FIG. 16, the producing method of the organic light-emitting device will be described. FIG. 16 shows schematic sectional views illustrating the producing method of the organic light-emitting device according to the embodiment 2-1 of the present invention.

25 (1) First, an anode 105 is formed on a substrate 106, as shown in FIG.

16(a) (Anode forming step).

(2) Then, a medium 104' is disposed over the substrate 106, as shown in FIG. 16(b). The medium 104' means the coating film that is produced by the application of the solution obtained by two kinds of material, i.e., 5 organic polymer and organic material that can be eluted to make the layer porous being dissolved in solvent (Medium disposing step).

(3) Then, the medium 104' is dried by heating or by airstream and treated by use of the solvent that does not allow solubilization of the organic polymer and allows solubilization of only the organic material, so as to elute 10 only the organic material, as shown in FIG. 16(c). Then, the region where the organic material are present is hollowed and thereby the organic polymer layer is made porous to thereby produce the organic layer 104 (Porosity producing step).

In the porosity producing step, at least a part of the organic layer 104, 15 to be more specific, at least one of a part of the organic layer 104 on the anode 105 side and a part of the same on the cathode 101 side can be made porous.

(4) Then, luminous molecules 103 are dispersed over the organic layer 104, as shown in FIG. 16(d) (Luminous molecule dispersing step).

20 (5) Then, a leveled layer 102 comprising the charge transport material is formed on the organic layer 104, as shown in FIG. 16(e) (Leveled layer forming step).

(6) Then, the cathode 101 is formed on the leveled layer 102, as shown in FIG. 16(f) (Cathode forming step).

25 The proportion of the organic material to be mixed with the organic

polymer is preferable 10·50%. This is because when the proportion of the organic material is less than 10%, the layer (organic layer) cannot fully be made porous, while on the other hand, when the proportion of the organic material is more than 50%, the concentration of the layer is lowered, so that 5 the charge transport capabilities of the entire layer is deteriorated and the stiffness and adhesion of the layer to the substrate is weakened. The proportion of the organic material is further preferably 20·30%.

The organic materials that may be chosen for use include oligomer produced with low molecular weight for wide selection of the solvent, in 10 addition to the organic polymer.

The dispersion of the luminous molecules is preferably performed in the evaporation method. The luminous molecules may be brought into solution so that they can be steamed. In the method for penetrating the luminous molecules into the porous organic layer, the luminous molecules 15 are preferably steamed after dispersed (In the embodiment 2·2 mentioned later as well, the luminous molecules are preferably steamed after dispersed).

The organic light-emitting device is enabled to take out the surface emission by making at least one electrode transparent or translucent. 20 Usually, the ITO (indium tin oxide) film is often used for the anode serving as the hole injection electrode. Tin oxide, Ni, Au, Pt and Pd can additionally be cited as the material therefor. The ITO film is formed by use of the deposition method, such as sputtering, electron beam evaporation and ion plating, for the purpose of improving the transparency or reducing 25 the resistivity.

The film thickness is determined from the required sheet resistance and visible light transmittance. The organic light-emitting device is relatively high in driving current density, so that it is often used with thickness of 100nm or more in order to reduce the sheet resistance.

5 An alloy comprising a metal of low work function and low electron injection barrier, such as MgAg alloy or AlLi alloy proposed by Tang et al., and a metal of relatively large work function and stable is often used for the cathode serving as the electron injection electrode. For the purpose of the metal of low work function being deposited on the organic layer side to  
10 protect the metal of low work function, the metal of large work function may be laminated thickly. Alternatively, a laminated electrode, such as Li/Al and LiF/Al, may be used therefor. These cathodes are preferably formed in the evaporation method or the sputtering method. When the electron injection material, such as dilithium phthalocyanine, disodium  
15 phthalocyanine, magnesium porphin, 4,4,8,8-tetrakis (1H-pyrazole-1-yl) pyrazabole, is used for the electrode, the electrode can be formed by only the metal of large work function and stable. This makes it resistant to reaction such as oxidizing and thus enables an enhanced life property.

The substrate may be formed of any material that can support the  
20 organic light-emitting device laminating the thin film thereon and is transparent or translucent to take out the emission generated in the organic layer. Glass such as Corning 1737 or polyester or other resin film may be used therefore.

FIG. 17 is a schematic sectional view showing a variant of the organic  
25 light-emitting device according to the embodiment 2-1 of the present

invention.

As shown in FIG. 17, the organic light-emitting device 107 has the constitution in which the luminous molecules 103 of the material contributable to the emission are penetrated in the porous organic layer 104 in the vicinity of the front surface. The luminous molecules 103 can be penetrated in the porous organic layer 104 in the vicinity of the front surface by steaming.

This constitution enables the luminous molecules 103 to be collected in the porous organic layer 104 in the vicinity of the front surface thereof and 10 also enables the surface area in the hole-and-electron recombination region in which the material contributable to the emission to be enlarged, thus enabling the emission with high luminance. The luminous molecules may be disposed on and in the vicinity of the front surface of the organic layer 104.

15 (Experimental Example 2-1)

Experimental Example 2-1 illustrates an actual example of the organic light-emitting device of the embodiment 2-1. In the following, the description thereon will be given.

Polyvinyl carbazol and butyral resin of low degree of polymerization 20 (available from Sekisui Chemical Co., Ltd., S-LEC B, Part number BL-S) were dissolved in toluene in the proportion 80:20 by weight, to produce the solution.

Then, the solution thus produced was deposited on the ITO-deposited glass substrate in the spin coat method to produce the organic layer having 25 thickness of 100nm.

The ITO substrate forming thereon the organic layer was dipped in N,N-dimethylformamide, so that only the butyral resin (S-LEC B) was dissolved and removed and, thereafter, it was dried at 200°C by heating, to thereby produce the porous organic layer.

5 After the substrate was cooled down to room temperature in a vacuum chamber, the coumarin 6 of laser dyes used as the luminous molecules were dispersed on it at an evaporation rate of 0.01nm/s for 10 seconds by resistance heating.

Sequentially, 4,4,8,8-tetrakis (1H-pyrazole-1-yl) pyrazabole used as the  
10 electron injection material was evaporated at the evaporation rate of 0.01nm/s for 1 minute.

Finally, an Al electrode was formed at the evaporation rate of 1nm/s to have layer thickness of about 100nm and thereby the organic light-emitting device.

15 When the section of the device was observed by SEM, it was found that the porous layer having diameter of the order of 3-6nm was formed and the porous region was filled with the electron injection material. When a direct current voltage was applied to this device for evaluation purpose, it was found that the green luminescence of coumarin 6 was obtained and glowed  
20 on stably with current efficiency of 8.0cd/A.

TABLE 2

	Current efficiency (cd/A)
Experimental Example 2-1	8.0
Experimental Example 2-2	8.2
Experimental Example 2-3	7.5
Experimental Example 2-4	3.2

## (Experimental Example 2-2)

In the organic light-emitting device of the experimental example 2-2, 4,4,8,8-tetrakis (1H-pyrazole-1-yl) pyrazabole was evaporated at the evaporation rate of 0.1nm/s for 2 minutes to thereby produce a leveled layer, for the purpose of providing the leveled layer, instead of filling the porous layer with the electron injection material, in the evaporation of the electron injection material of the experimental example 2-1.

When the section of the device was observed by SEM, it was found that the leveled layer was formed on the porous layer having diameter of the order of 3-6nm. When a direct current voltage was applied to this device for evaluation purpose, it was found that the green luminescence of coumarin 6 was obtained and glowed on stably with current efficiency of 8.2cd/A.

## 15 (Experimental Example 2-4)

The experimental example 2-4 is for comparison with the experimental examples 2-1 and 2-2 (the organic layer is not made porous).

Polyvinyl carbazol, 2-(4-biphenyl)-5-(4-t butylphenyl)-1,3,4-oxadiazole used as the electron transport material, and coumarin 6 used as the luminous molecule material were dissolved in a weight ratio of 100:40:0.2 in the mixed solvent with a ratio of toluene to THF of 1:1 to produce the silution.

Thereafter, the solution was applied to the ITO deposited glass substrate by use of the spinner, to form the organic layer having thickness of 25 100nm.

The cathode comprising Li and Al of 1nm used as the electron injection electrode was formed on that organic layer, to thereby produce the organic light-emitting device.

When a direct current voltage was applied to this device for evaluation 5 purpose, it was found that the green luminescence of coumarin 6 was obtained and the current efficiency was 3.2cd/A, as shown in TABLE 2.

Thus, the organic light-emitting device having the organic layer as was made porous was dramatically improved in current efficiency, as compared with the one having the organic layer as was not made porous.

10 [Embodiment 2-2]

Embodiment 2-2 relates to the organic light-emitting device having the structure in which the emission region is roughened. FIG. 18 is a schematic sectional view of an organic light-emitting device according to the embodiment 2-2 of the present invention

15 As shown in FIG. 18, an organic light-emitting device 110 comprises the anode 105, the cathode 101 arranged in opposition to the anode 105, an emission region 113 arranged between the anode 105 and the cathode 101, and a leveled layer arranged between the cathode 101 and the emission region 113.

20 The emission region 113 comprises an organic layer 120 which is made porous on the cathode 101 side thereof, and the luminance molecules 103 arranged on the roughened front surface of the organic layer 120.

Further, the leveled layer 102 comprising charge transport material is formed on the organic layer 120 and the cathode 101 is laminated on the 25 leveled layer 102.

This constitution, in which the luminous molecules 103 are collected on the emission region 113 (on the front surface of the organic layer 120) which is roughened to enlarge its surface area and also the hole-and-electron recombination region in which the luminous molecules 5 103 are present is roughened to enlarge its surface area, can produce the emission with increased luminance. It is to be noted that as is the case with the embodiment 2-1, the luminous molecules may be penetrated in the organic layer in the vicinity of the front surface thereof or may alternatively be arranged on as well as in the vicinity of the front surface of the organic 10 layer.

Referring now to FIG. 19, the producing method of the organic light-emitting device will be described. FIG. 19 shows schematic sectional views illustrating the producing method of the organic light-emitting device according to the embodiment 2-2 of the present invention.

15 (1) The anode 105 is formed on the substrate 106 in the same manner as in the step (1) of the producing method as illustrated in Embodiment 2-1 (FIG. 19(a)).

(2) Then, a coating film 120' of a medium is formed on the anode 105 by use of organic polymer forming the organic layer (FIG. 19(b)).

20 (3) Then, the coating film 120' is etched by dry etching using e.g. a reactive ion etching (RIE) to roughen the front surface of the coating film 120' so as to form the organic layer 120 (FIG. 19(c)).

(4) Then, the luminous molecules 103 are dispersed on the roughened front surface of the organic layer 120 (FIG. 19(d)). The dry etching may be 25 performed by use of a general purpose etching device such as a barrel type

one and a parallel flat-plate type one. Depending on the state of the organic layer, Ar gas may simultaneously be introduced.

(5) Then, the leveled layer 102 comprising the charge transport material is formed on the organic layer 120 in the same manner as in 5 Embodiment 2-1 (FIG. 19(e)).

(6) Then, the cathode 101 is formed on the leveled layer 102 (FIG. 19(f)).

(Experimental Example 2-3)

Experimental Example 2-3 shows an actual example of the organic 10 light-emitting device of Embodiment 2-2. Specifically, the solution in which polyvinyl carbazol was dissolved in toluene was deposited on the ITO-deposited glass substrate in the spin coat method to produce the organic layer having thickness of 100nm.

Then, the ITO substrate forming thereon the organic layer was 15 roughened in the dry etching device of parallel flat-plate type in the conditions of: an oxygen flow rate of 60sccm; pressure of 40mTorr; and high-frequency output power of 100W for 1 minute and then was disposed in the vacuum chamber.

Then, the coumarin 6 of laser dyes used as the luminous molecules 20 were dispersed on the substrate at an evaporation rate of 0.01nm/s for 10 seconds in the vacuum evaporation method using the resistance heating.

Sequentially, 4,4,8,8-tetrakis (1H-pyrazole-1-yl) pyrazabole used as the electron injection material was evaporated at the evaporation rate of 0.1nm/s for 2 minutes.

25 Finally, the Al electrode was formed at the evaporation rate of 1nm/s to

have layer thickness of about 100nm and thereby the organic light-emitting device.

When the section of the device was observed by SEM, it was found that the roughened surface of the order of  $\pm 3\text{nm}$  and the leveled layer formed by 5 the electron injection material were formed in the organic layer. When a direct current voltage was applied to this device for evaluation purpose, it was found that the green luminescence of coumarin 6 was obtained and glowed on stably with current efficiency of 7.5cd/A, as shown in TABLE 2.

Thus, the organic light-emitting device shown in Experimental 10 example 2-3 was dramatically improved in current efficiency by roughening the organic layer, as compared with the organic light-emitting device shown in Experimental example 2-4.

[Embodiment 2-3]

While Embodiments 2-1 and 2-2 relate to the organic light-emitting 15 devices having the structure in which the emission region is made porous or roughened to produce improved luminous efficiency, this embodiment 2-3 relates to the organic light-emitting device having the structure in which the charge transport region is roughened. FIG. 20 is a schematic sectional view showing an organic light-emitting device according to an embodiment 20 2-3 of the present invention.

As shown in FIG. 20, an organic light-emitting device 115 comprises the anode 105, the cathode 101 arranged in opposition to the anode 105, an emission region 117 arranged between the anode 105 and the cathode 101, and a charge transport layer 116 arranged between the emission region 117 25 and the anode 105.

The charge transport layer 116 is roughened at the emission region 117 side by the dry etching. In the structure above, the charge transport layer 116 is the hole transport layer.

This constitution can provide an enhanced contact area of the emission region 117 and the charge transport region 116 and an improved injection efficiency of the holes injected from the anode 105 to the emission region 117.

While in this embodiment, reference is given to the constitution of the charge transport region being roughened, it is needless to say that the charge transport region may be made porous to achieve the equivalent effect to the effect produced by roughening the charge transport region. When the charge transport layer 116 has luminescent properties, the emission region 117 is not necessarily needed, but instead at least either of the anode 105 side and the cathode 101 side of the charge transport layer 116 may be made porous or roughened to produce an improved injection efficiency of the charge from the electrode, as is the case with the above.

### Industrial Applicability

As mentioned above, the constitution of the present invention can attain the objects of the present invention satisfactorily.

According to the invention of the first inventive group, the luminous molecules, or the luminous molecules and the charge transport material are penetrated in the polymer, or the polymer in which the charge transport material is dispersed, to thereby provide the organic light-emitting device capable of producing an improved luminous efficiency and at the same time

facilitating the patterning even in the polymer dispersed organic light-emitting device.

According to the invention of the second inventive group, in the polymer-based organic light-emitting device, the emission region, which was commonly diffused over the whole organic layer, is collected to a specific region and also the hole-and-electron recombination region where the luminous molecules are present is made porous or roughened to enlarge the surface area, to thereby produce an improved luminous efficiency.

**What is claimed is:**

1. A light-emitting device having an emission region between an anode and a cathode,
  - 5 wherein the emission region comprises material contributable to emission and a medium for containing the material, and  
wherein the material contributable to the emission has a substantially successive distribution of concentration from the anode side of the emission region toward the cathode side thereof.
- 10 2. The light-emitting device as set forth in Claim 1, wherein the material contributable to the emission exhibits the distribution of concentration according to which any one of the anode side of the emission region and the cathode side thereof is higher in concentration than the other, and the  
15 concentration reduces from the one side toward the other side successively.
3. The light-emitting device as set forth in Claim 1, wherein the emission region further comprises charge transport material.
- 20 4. The light-emitting device as set forth in Claim 3, wherein the charge transport material has a substantially successive distribution of concentration from the anode side of the emission region toward the cathode side thereof.
- 25 5. A light-emitting device having a charge transport region between an

anode and a cathode,

wherein the charge transport region comprises charge transport material and a medium for containing the charge transport material, and

wherein the charge transport material has a substantially successive distribution of concentration from the cathode side of the charge transport region toward the anode side thereof.

6. The light-emitting device as set forth in Claim 1, wherein the emission region includes a region where the material contributable to the emission is

10 not present.

7. The light-emitting device as set forth in Claim 1, wherein a part of the emission region that exhibits the maximum concentration of the material contributable to the emission is away from the anode and the cathode.

15

8. The light-emitting device as set forth in Claim 5, wherein the charge transport region includes a region where the charge transport material is not present.

20 9. The light-emitting device as set forth in Claim 5, wherein a part of the charge transport region that exhibits the maximum concentration of the charge transport material is away from the anode and the cathode.

10. A lighting device using the light-emitting device as set forth in Claim 1.

25

11. A light-emitting device having an emission region between an anode and a cathode,
- wherein the emission region comprises material contributable to emission and a medium for containing the material, and
- 5       wherein the material contributable to the emission has a distribution of concentration that reduces substantially successively in a direction parallel to a surface of the cathode and a surface of the anode from a substantially center of the emission region toward a periphery thereof.
- 10      12. The light-emitting device as set forth in Claim 11, wherein there are provided a number of materials contributable to the emissions and are adjacently arranged in a direction parallel to a surface of the cathode and a surface of the anode, and wherein the materials contributable to the emissions are different from each other in luminous color.
- 15
13. The light-emitting device as set forth in Claim 11, wherein the emission region further comprises charge transport material.
14. The light-emitting device as set forth in Claim 13, wherein the charge
- 20     transport material has a distribution of concentration that reduces in the direction parallel to the surface of the cathode and the surface of the anode from the substantially center of the emission region toward the periphery thereof.
- 25    15. The light-emitting device as set forth in Claim 11, wherein the

material contributable to the emission has a substantially successive distribution of concentration from the cathode side of the emission region toward the anode side thereof.

5     16. The light-emitting device as set forth in Claim 14, wherein the charge transport material has a substantially successive distribution of concentration from the cathode side of the emission region toward the anode side thereof.

10    17. The light-emitting device as set forth in Claim 11, wherein the emission region includes a region where the material contributable to the emission is not present.

15    18. The light-emitting device as set forth in Claim 11, wherein the medium for containing the material contributable to the emission has charge transport capabilities.

20    19. The light-emitting device as set forth in Claim 11, wherein the medium for containing the material contributable to the emission comprises organic material.

25    20. The light-emitting device as set forth in Claim 11, wherein the medium comprises polymer.

25    21. A display using the light-emitting device as set forth in Claim 11.

22. A producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising:
- 5        a disposing step of disposing a medium on the anode or the cathode;
- and
- 10      a containing step of allowing material contributable to emission to be contained in the medium to form the emission region.
23. A producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising:
- 15      a disposing step of disposing a medium containing charge transport material on the anode or the cathode; and
- 20      a containing step of allowing material contributable to emission to be contained in the medium to form the emission region.
24. A producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising:
- 25      a disposing step of disposing a medium on the anode or the cathode;
- and
- 20      a containing step of allowing material contributable to emission and charge transport material to be contained in the medium.
25. A producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising:
- 25      a disposing step of disposing a medium containing charge transport

material on the anode or the cathode; and  
a containing step of allowing material contributable to emission and  
charge transport material to be contained in the medium.

5       26. A producing method of a light-emitting device having a charge  
transport region between an anode and a cathode, the producing method  
comprising:

            a disposing step of disposing a medium on the anode or the cathode;  
and

10       a containing step of allowing charge transport material to be contained  
in the medium.

15       27. The producing method of a light-emitting device as set forth in Claim  
23, wherein in the containing step, the material contributable to the  
emission is penetrated into the medium, whereby it is contained in the  
medium.

20       28. The producing method of a light-emitting device as set forth in Claim  
24, wherein in the containing step, the material contributable to the  
emission and the charge transport material are penetrated into the medium,  
whereby they are contained in the medium.

25       29. The producing method of a light-emitting device as set forth in Claim  
23, wherein in the containing step, solution obtained by the material  
contributable to the emission being dissolved in solvent is brought into

contact with the medium, whereby the material is penetrated into the medium.

30. The producing method of a light-emitting device as set forth in Claim  
5 24, wherein in the containing step, solution obtained by the material  
contributable to the emission and the charge transport material being  
dissolved in solvent is brought into contact with the medium, whereby the  
materials are penetrated into the medium.

10 31. The producing method of a light-emitting device as set forth in Claim  
27, wherein in the containing step, the material contributable to the  
emission is penetrated into the medium in an ink jet method.

15 32. A light-emitting device having an emission region between an anode  
and a cathode,

wherein the emission region comprises material contributable to  
emission, and

20 wherein a collecting means for collecting the material contributable to  
the emission to a specific region is provided between the anode and the  
cathode.

33. A light-emitting device having an emission region between an anode  
and a cathode,

25 wherein at least one of an anode side of the emission region and a  
cathode side thereof is made porous, and

wherein material contributable to emission is included in a surface of the emission region which is made porous.

34. A light-emitting device having an emission region between an anode  
5 and a cathode,

wherein at least one of an anode side of the emission region and a cathode side thereof is made porous, and

wherein material contributable to emission is included in a region in the vicinity of a surface of the emission region which is made porous.

10

35. The light-emitting device as set forth in Claim 33, wherein charge transport material is included in a surface of the emission region which is made porous.

15

36. The light-emitting device as set forth in Claim 33, wherein a leveled layer comprising charge transport material is provided on a surface of the emission region which is made porous.

20

37. A light-emitting device having a charge transport region between an anode and a cathode,

wherein at least one of an anode side of the charge transport region and a cathode side thereof is made porous.

25

38. The light-emitting device as set forth in Claim 37, wherein the charge transport region is a hole transport region.

39. The light-emitting device as set forth in Claim 37, wherein the charge transport region is an electron transport region.

5 40. The light-emitting device as set forth in Claim 33, wherein the emission region comprises an organic matter.

41. The light-emitting device as set forth in Claim 33, wherein the emission region comprises polymer.

10

42. A light-emitting device having an emission region between an anode and a cathode,

wherein at least one of an anode side of the emission region and a cathode side thereof is roughened, and

15

wherein material contributable to emission is included in a surface of the roughened emission region.

43. A light-emitting device having an emission region between an anode and a cathode,

20

wherein at least one of an anode side of the emission region and a cathode side thereof is roughened, and

wherein material contributable to emission is included in a region in the vicinity of a roughened surface of the emission region.

25

44. The light-emitting device as set forth in Claim 42, wherein a leveled

layer comprising charge transport material is provided on a roughened surface of the emission region.

45. A light-emitting device having a charge transport region between an  
5 anode and a cathode,

wherein at least one of an anode side of the charge transport region and a cathode side thereof is roughened.

46. The light-emitting device as set forth in Claim 45, wherein the charge  
10 transport region is a hole transport region.

47. The light-emitting device as set forth in Claim 45, wherein the charge transport region is an electron transport region.

15 48. The light-emitting device as set forth in Claim 42, wherein the emission region comprises an organic matter.

49. The light-emitting device as set forth in Claim 42, wherein the emission region comprises polymer.

20 50. A display using the light-emitting device as set forth in Claim 33.

51. A lighting device using the light-emitting device as set forth in Claim 33.

52. A producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising:

a medium disposing step of disposing a medium on the anode or the cathode; and

5        a porosity producing step of making at least a part of the medium porous.

53. A producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising:

10        a medium disposing step of disposing a medium on the anode or the cathode;

      a porosity producing step of making at least one of an anode side of the medium and a cathode side thereof porous; and

15        a disposing step of disposing material contributable to emission on a porous surface of the medium, so that the emission region is formed by the medium and the material contributable to the emission.

54. A producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising:

20        a medium disposing step of disposing a medium on the anode or the cathode;

      a porosity producing step of making at least one of an anode side of the medium and a cathode side thereof porous;

25        a containing step of allowing material contributable to emission to be contained in a region in the vicinity of a porous surface of the medium, so

that the emission region is formed by the medium and the material contributable to the emission; and

a disposing step of disposing charge transport material on a porous surface of the medium.

5

55. The producing method of a light-emitting device as set forth in Claim 53, which comprises a disposing step of disposing charge transport material on a porous surface of the emission region.

10 56. The producing method of a light-emitting device as set forth in Claim 53, which comprises a leveled layer forming step of forming a leveled layer comprising charge transport material on the emission region.

15 57. The producing method of a light-emitting device as set forth in Claim 53, wherein the disposing step is a step of disposing a medium containing material soluble in a specified solvent, and the porosity producing step is a step of eluting the material from the solvent to thereby make the medium porous.

20 58. A producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising:

a medium disposing step of disposing a medium on the anode or the cathode; and

a roughening step of roughening a part of the medium.

59. A producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising:

a medium disposing step of disposing a medium on the anode or the cathode;

5 a roughening step of roughening at least one of an anode side of the medium and a cathode side thereof; and

a disposing step of disposing material contributable to emission on a roughened surface of the medium, so that the emission region is formed by the medium and the material contributable to the emission.

10

60. A producing method of a light-emitting device having an emission region between an anode and a cathode, the producing method comprising:

a medium disposing step of disposing a medium on the anode or the cathode;

15 a roughening step of roughening at least one of an anode side of the medium and a cathode side thereof; and

a containing step of allowing material contributable to emission to be contained in a region in the vicinity of a roughened surface of the medium, so that the emission region is formed by the medium and the material contributable to the emission.

20

61. The producing method of a light-emitting device as set forth in Claim 59, which comprises a leveled layer forming step of forming a leveled layer comprising charge transport material on the emission region.

25

62. The producing method of a light-emitting device as set forth in Claim 59, wherein the roughening step is a step of roughening the emission region by dry etching.

**Abstract of the Disclosure**

An organic light-emitting device 10 has the constitution in which an emission region 3 between an anode 2 and a cathode 4 is laminated on a substrate 1. The emission region 3 includes polymer 3A, luminous molecules 3G which are material contributable to emission, and charge transport material 3F.

The luminous molecules 3G and the charge transport material 3F are high in concentration at a side close to the cathode 4 and low in concentration at a side close to the anode 2 in a layer thickness direction of the emission region 3. The organic light-emitting device can be formed by luminous material, or luminous material and charge transport material being penetrated into polymer, or polymer in which charge transport material is dispersed, thus providing highly improved luminous efficiency and at the same time facilitating the patterning.

FIG. 1

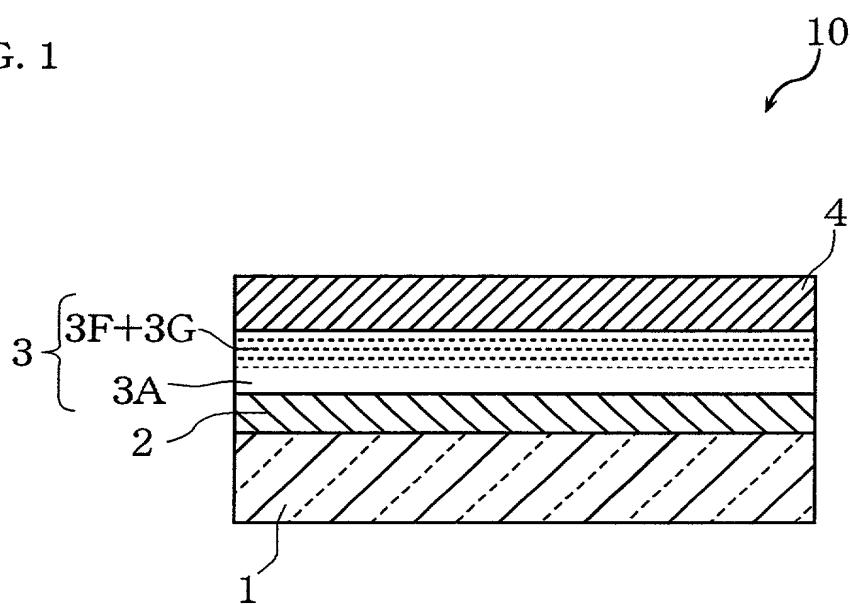
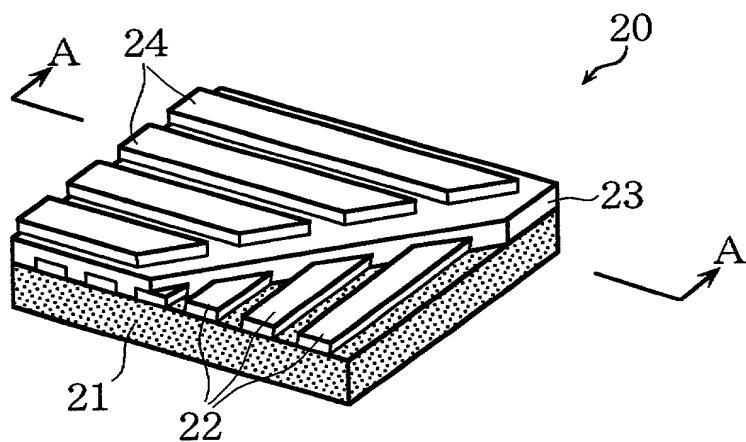


FIG. 2

(a)



(b)

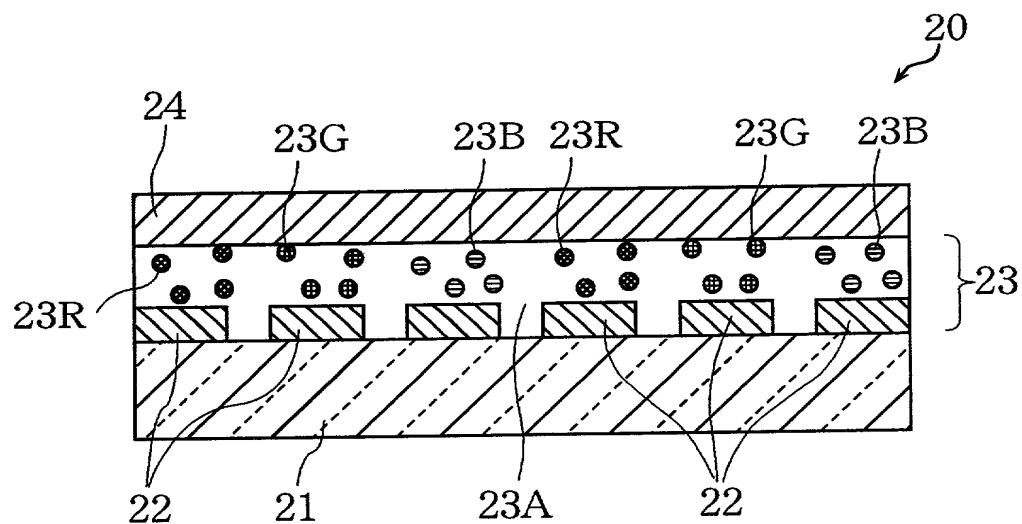


FIG. 3

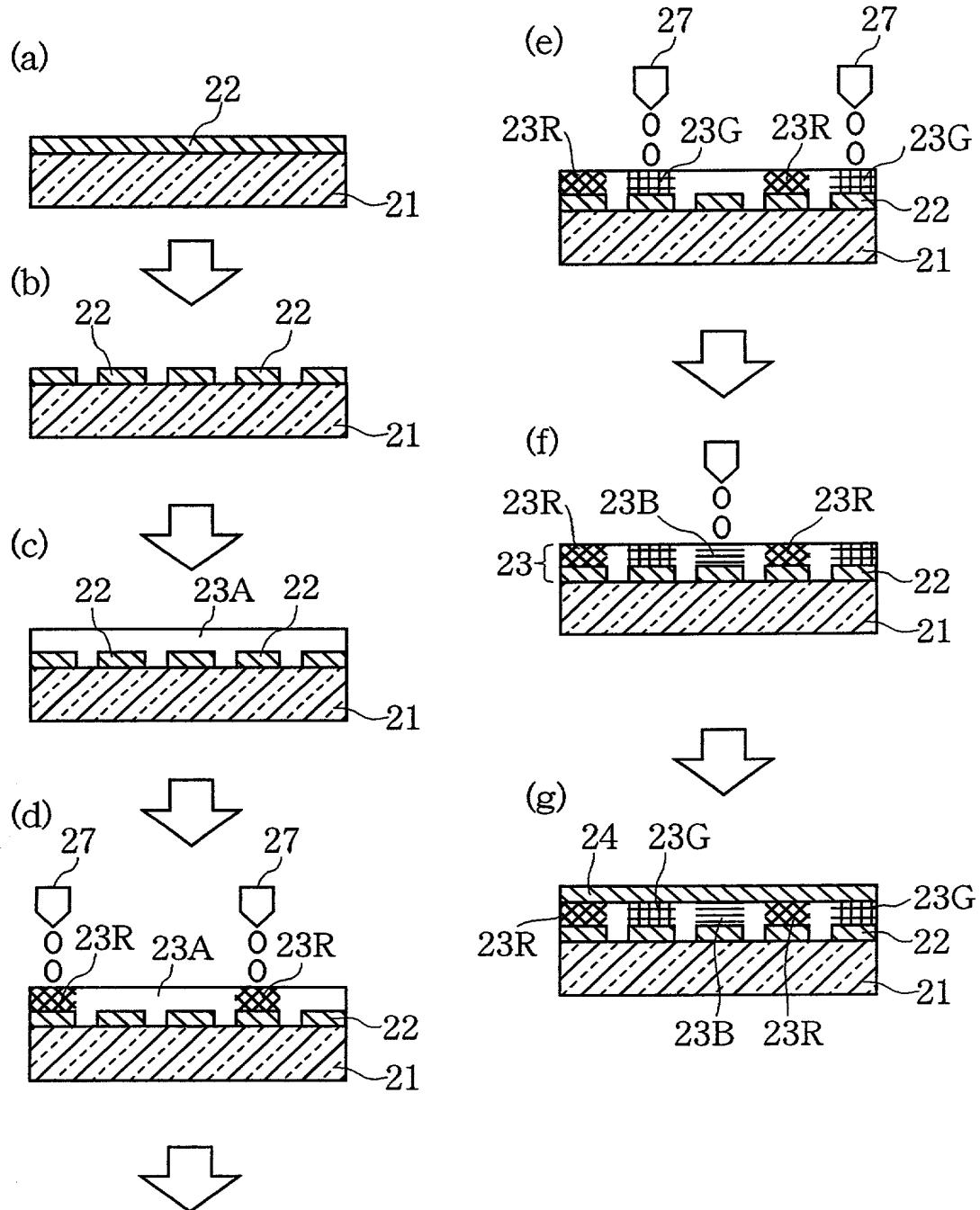


FIG. 4

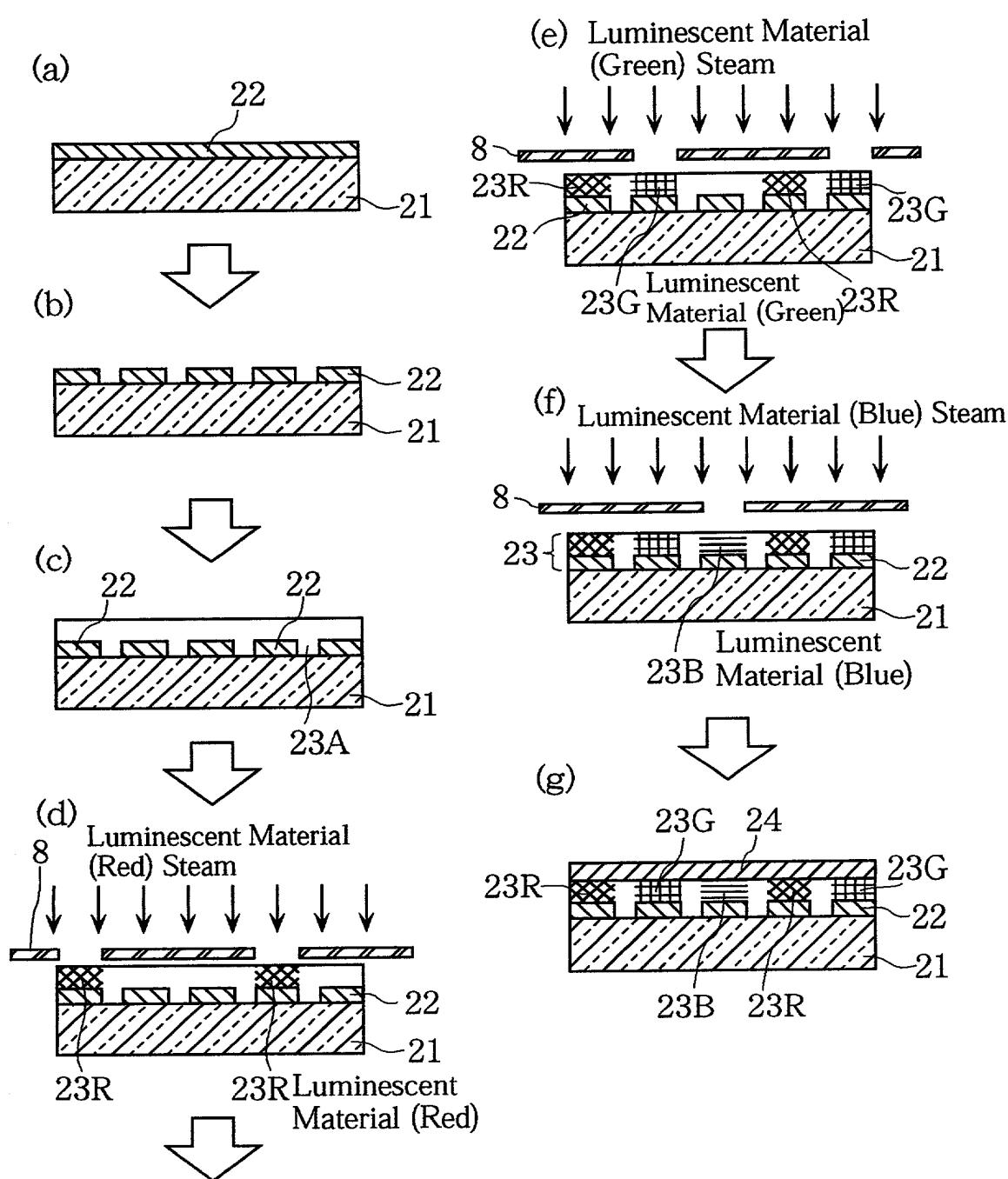


FIG. 5

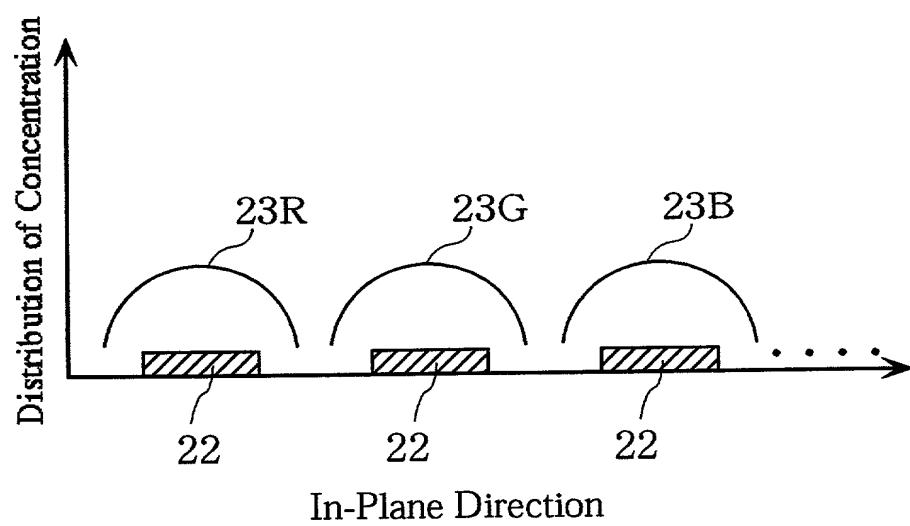


FIG. 6

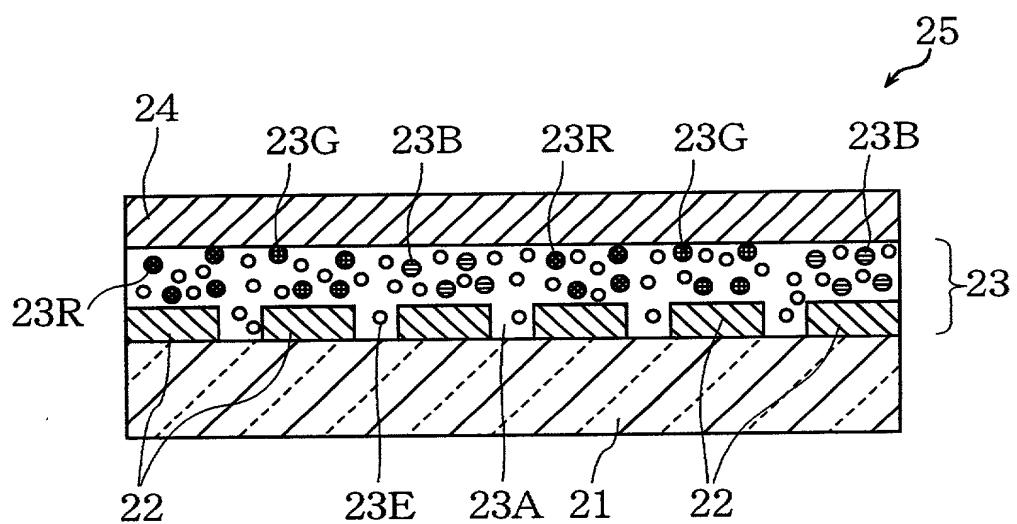


FIG. 7

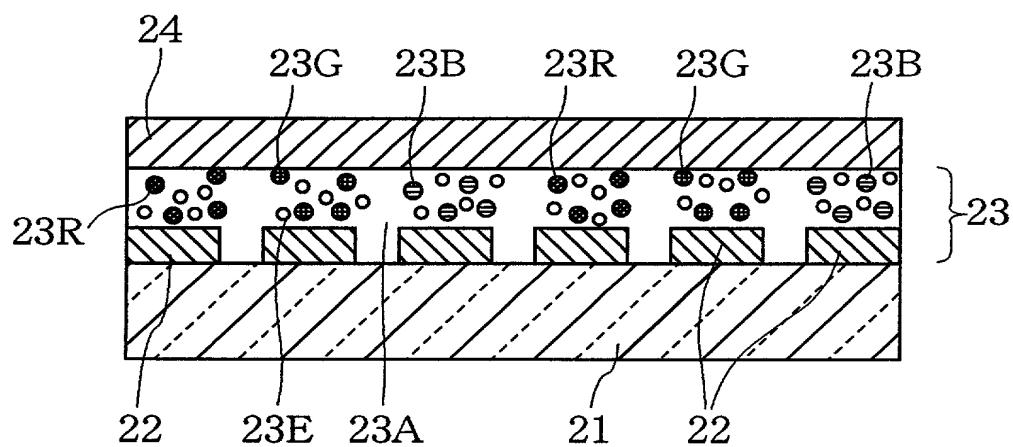


FIG. 8

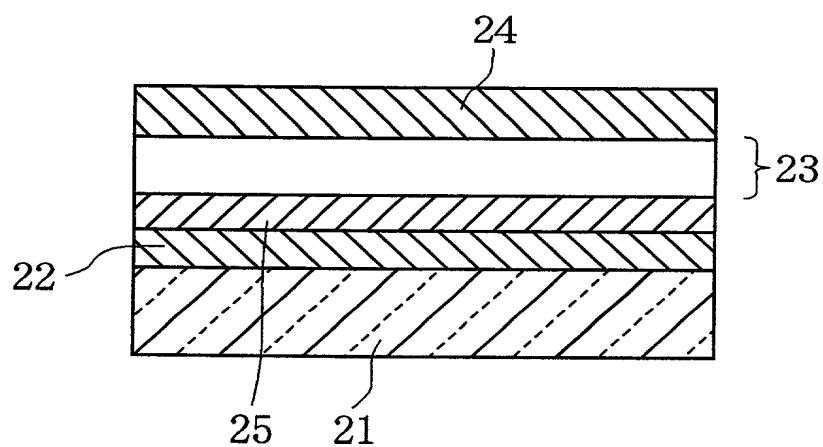


FIG. 9

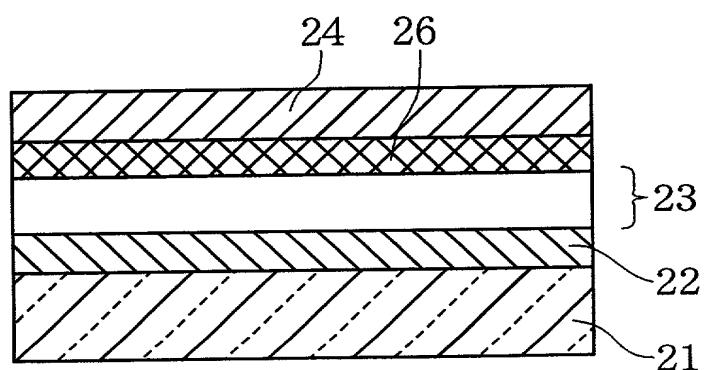


FIG. 10

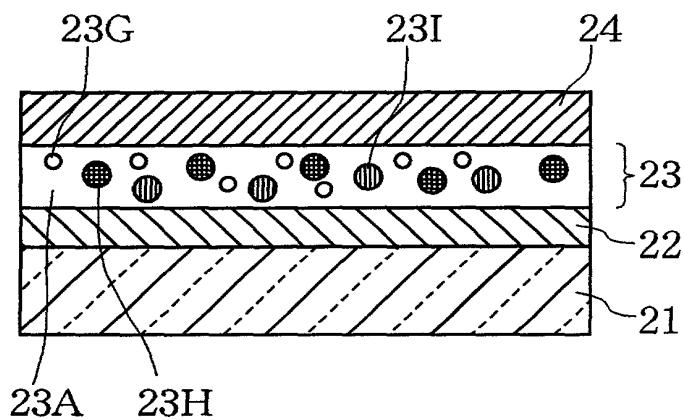


FIG. 11

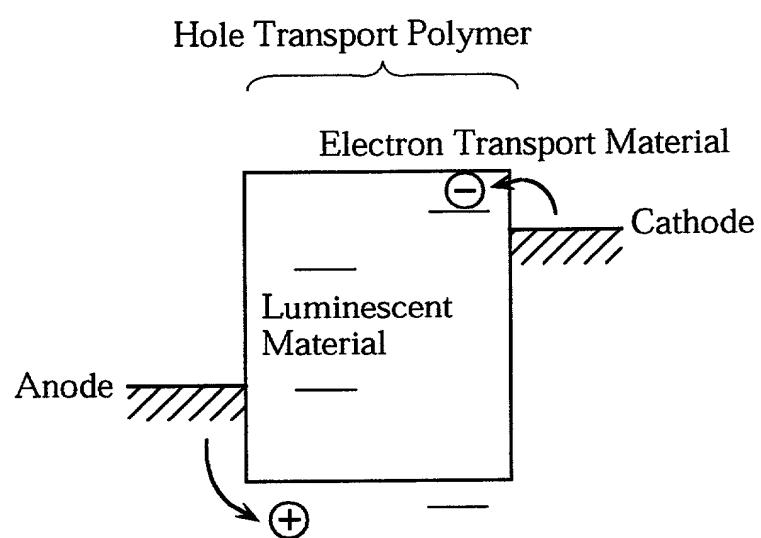


FIG. 12

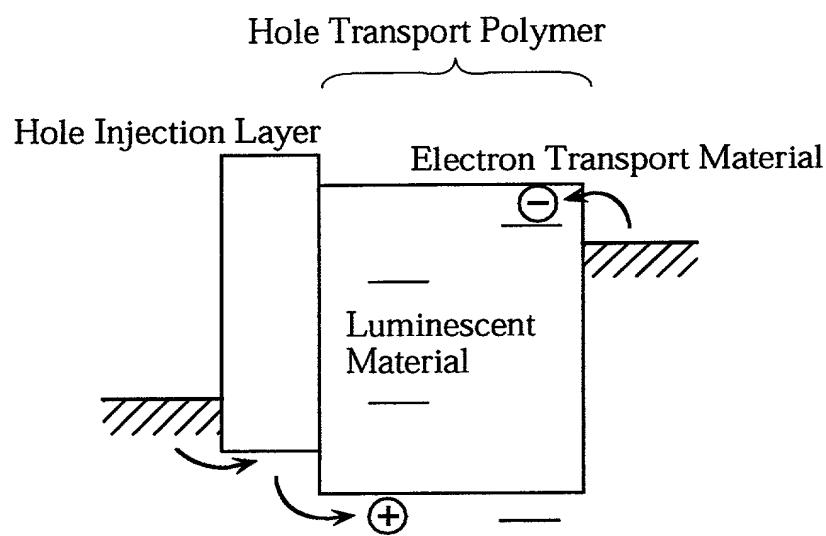


FIG. 13

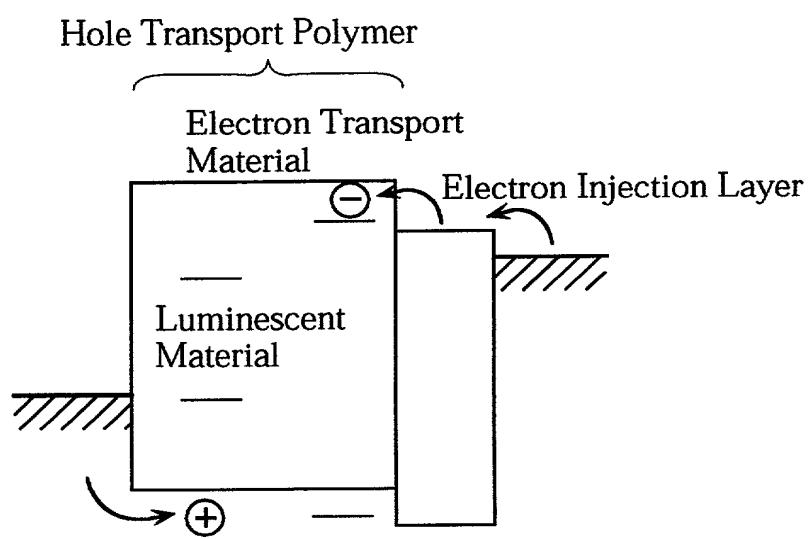


FIG. 14

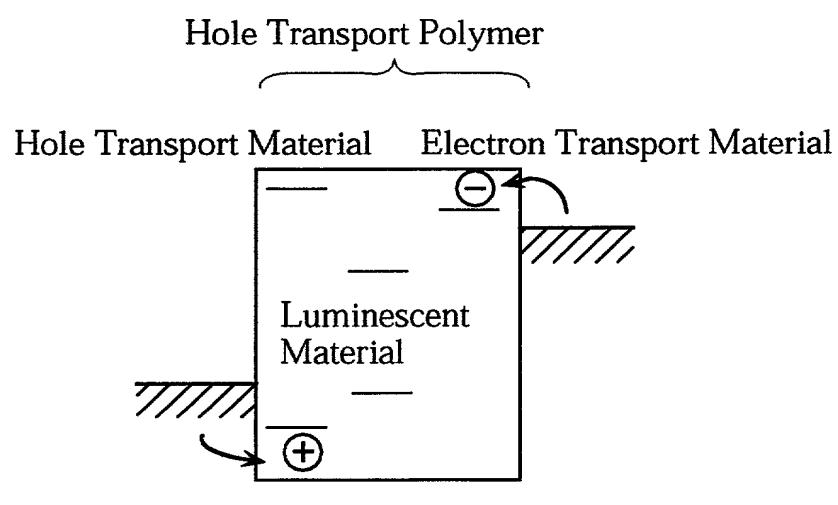


FIG. 15

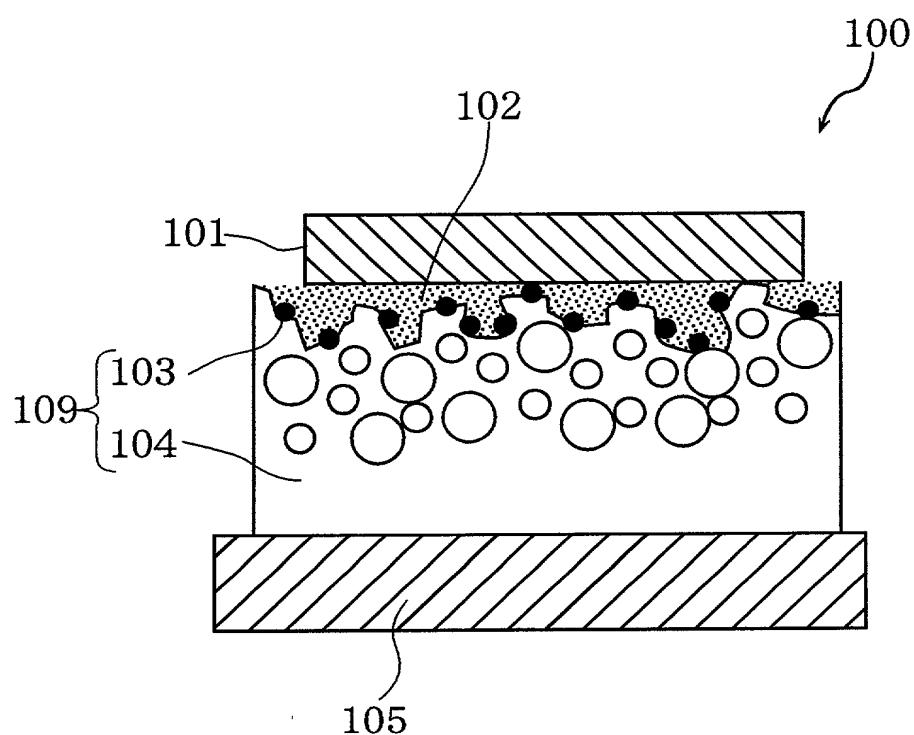


FIG. 16

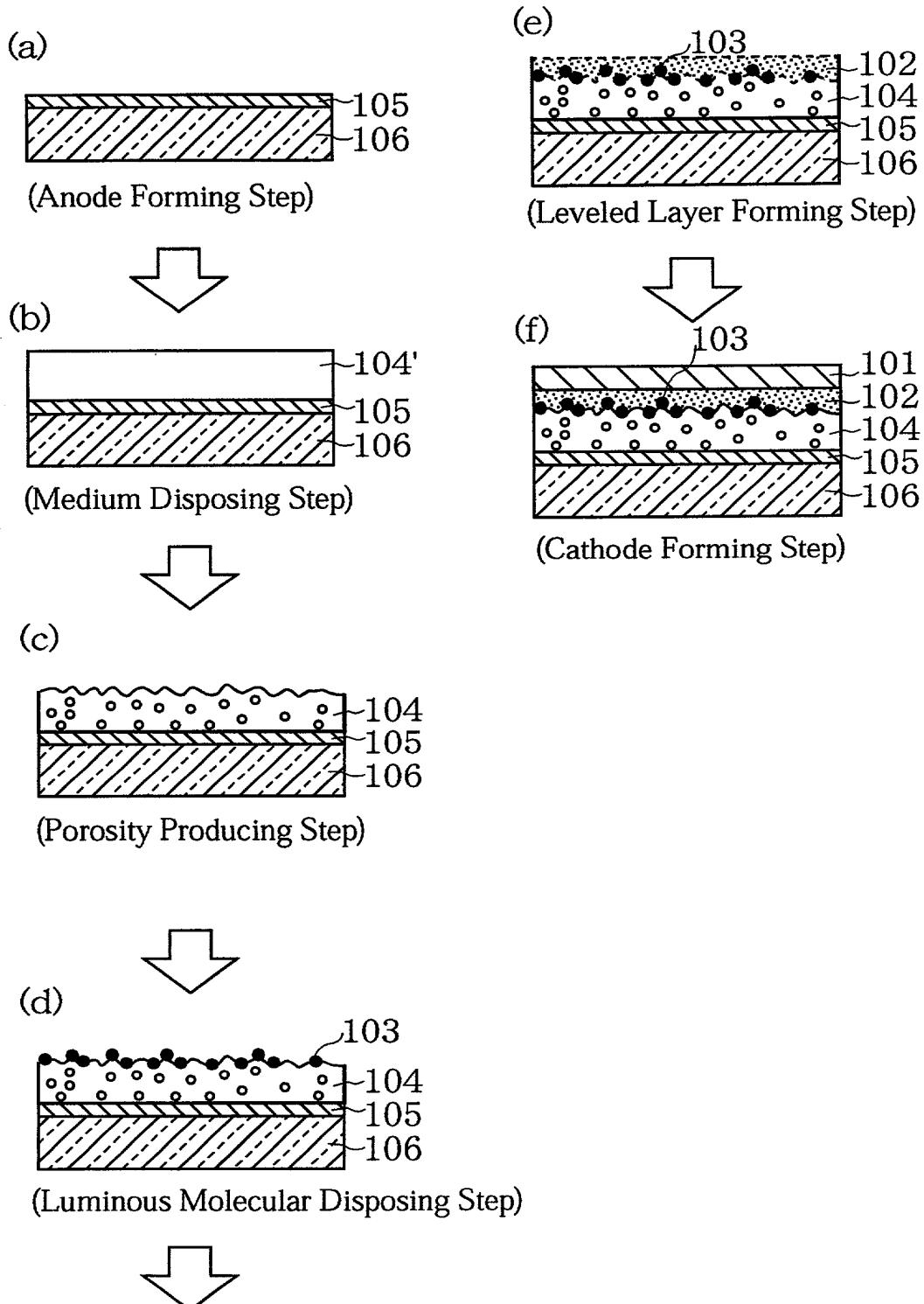


FIG. 17

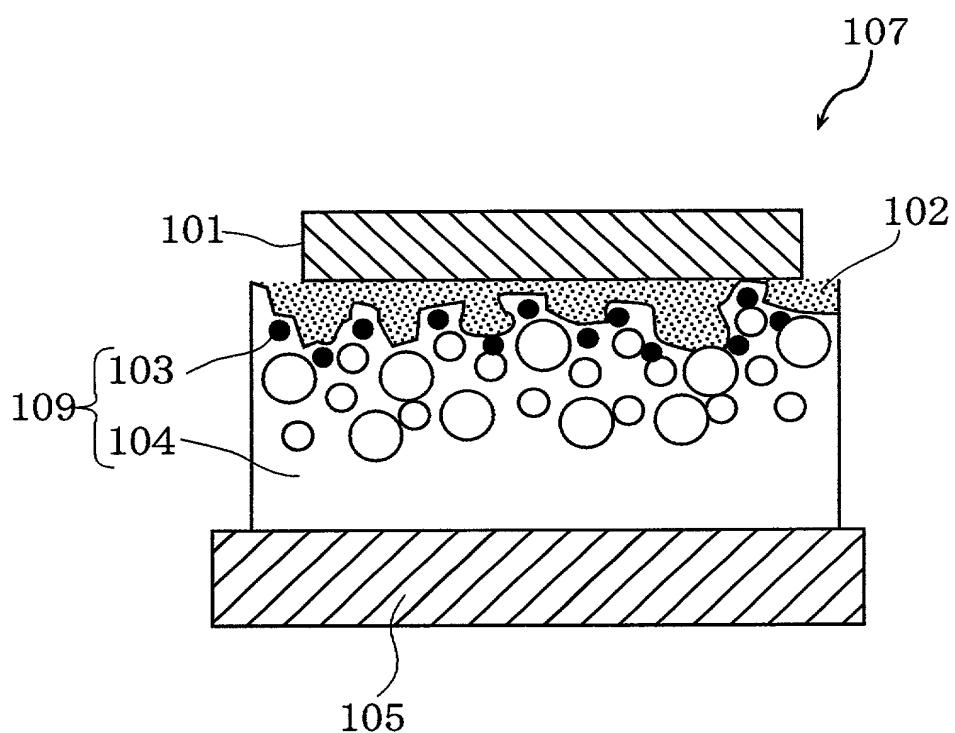


FIG. 18

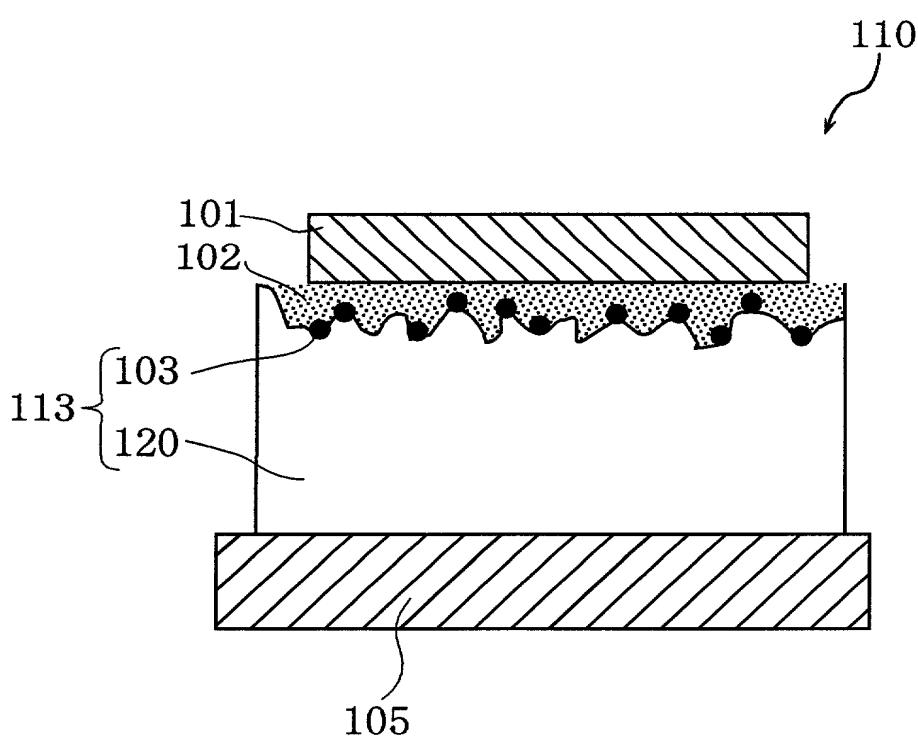


FIG. 19

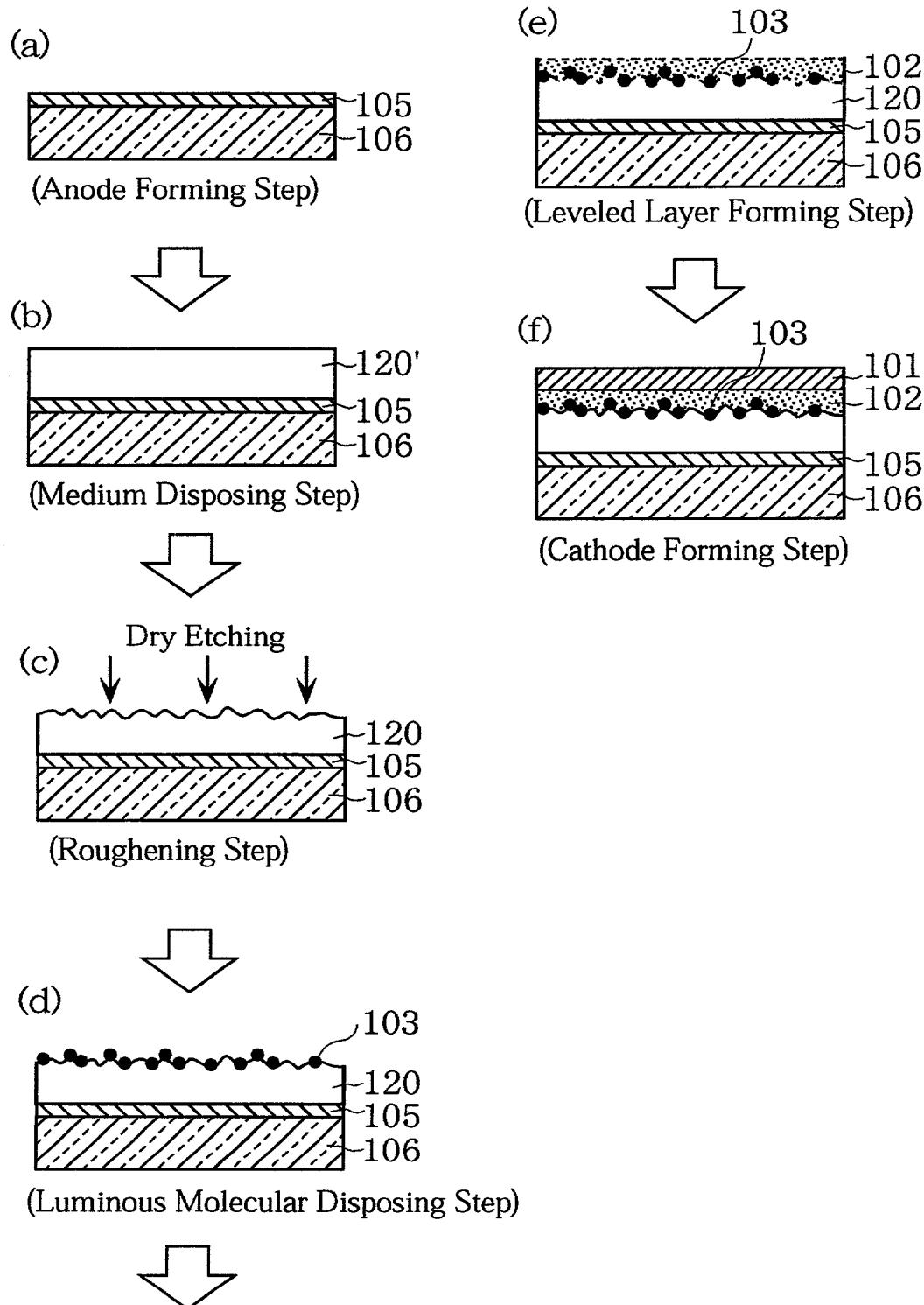
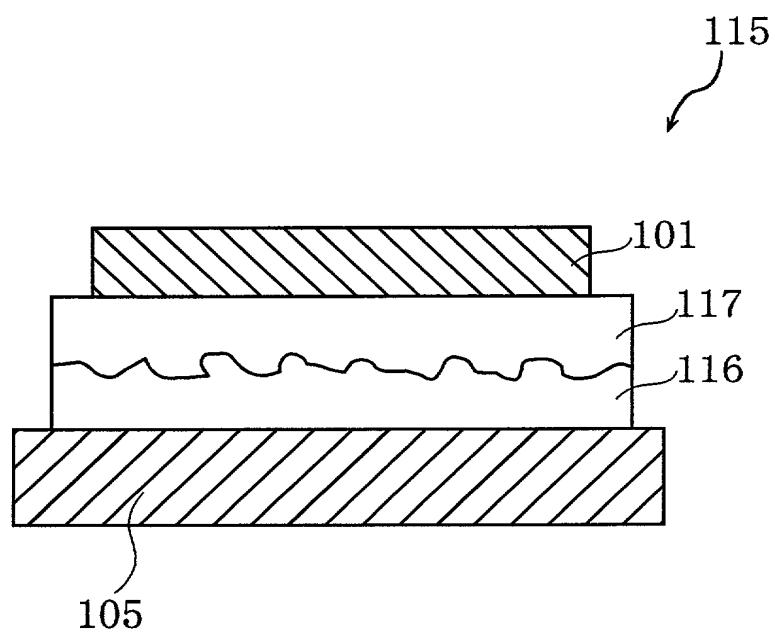


FIG. 20



**Declaration and Power of Attorney  
Under Patent Cooperation Treaty  
35 USC §371(c)(4)**

As a below named inventor, I hereby declare that:

my residence, post office address and citizenship are as stated below next to my name; that

I verily believe that I am the original, first and sole inventor (if only one name is listed below) or a joint inventor (if plural names are named below) of the invention entitled: LIGHT EMITTING ELEMENT AND PRODUCING METHOD THEREOF, AND DISPLAY DEVICE AND LIGHTING DEVICE USING THE SAME described and claimed in the international application number PCT/JP00/06971 filed October 5, 2000 and as amended on \_\_\_\_\_ (if any), the specification and claims of which I have reviewed and understand and for which I solicit a patent.

I acknowledge my duty to disclose information of which I am aware which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, §1.56(a), and that no application for patent or inventor's certificate on this invention has been filed in any country foreign to the United States of America prior to my international application by me or my legal representatives or assigns, except as follows:

Japanese Patent Application No. H11-284360 filed on October 5, 1999

Japanese Patent Application No. H11-299657 filed on October 21, 1999

The priority of the above applications (if any), filed within a year prior to my international application is hereby claimed under 35 USC 119. I hereby appoint the following as my attorneys of record with full power of substitution and revocation to prosecute this application and to transact all business in the patent office:

Roger W. Parkhurst, Reg. No. 25,177; Charles A. Wendel, Reg. No. 24,453; Lawrence D. Eisen, Reg. No. 41,009.

**ALL CORRESPONDENCE IN CONNECTION WITH THIS APPLICATION SHOULD BE SENT TO:  
PARKHURST & WENDEL, L.L.P., 1421 PRINCE STREET, SUITE 210, ALEXANDRIA, VIRGINIA 22314-2805, TELEPHONE (703) 739-0220.**

I hereby declare that I have reviewed and understand the contents of this Declaration, and that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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	Date of Signature	<u>April 23rd 2001</u>		
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	City	State or Province	Country	
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\*IF THERE IS MORE THAN ONE INVENTOR USE PAGE 2 AND PLACE AN "X" HERE

**PAGE 2 OF U.S.A. DECLARATION FORM**  
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*Apr. 23rd 2001*

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 address, including country)

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\*\*This form may be executed only when attached to the first page of the Declaration and Power of Attorney form and the specification (including claims) of the application to which it pertains.